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1 Introduction

Semiconductor quantum dot (QD) heterostructures have received considerable attention in the overall field of nanostructure research [1, 2]. One of the most studied systems are self-assembled InAs/GaAs QD's grown by molecular beam epitaxy in the Stransky-Krastanov mode. Transition of the growth mode from layer-by-layer growth to three dimensional islands occurs for an average thickness of 1–2 monolayers of InAs on GaAs. This results in the formation of InAs dots with a base size of the order of 10–20 nm on the top of an ultrathin InAs wetting layer. Because of the small size of the InAs dots, quantum confinement of the carriers occurs in all three spatial directions. This leads to atomic-like discrete states so that QD's are often described as artificial atoms. Along with the fundamental investigation of the physics of such structures, it is proposed that QD's be used as a means of fabricating nanostructures for optoelectronic applications, in particular for low threshold lasers [3], novel single-electron devices [4], optical storage devices [5, 6], photoconductive detectors [7], and – most recently – for quantum information devices [8].

Theoretical modelling of the optical properties of an ensemble of quantum dots is usually based on a set of rate equations, which describe the kinetics of the carrier exchange between the quantum dots and the barrier via the wetting layer [9, 10]. It is usually assumed that the carriers behave as correlated electron-hole pairs (excitons) during the thermally stimulated redistribution processes. Such excitonic models, which are widely used in the interpretation of experimental data for temperature-dependent optical properties, are attractive possibly because of their relative simplicity [11, 12]. However, so far, there is no direct experimental evidence for this assumption. We have recently suggested an alternative model for the

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photoluminescence (PL) in QD heterostructures [13] in which we considered the role of independent electrons and holes. In this paper we analyze results of two alterative models emphasizing the consequences of a difference in treatment of independent (uncorrelated) charge carriers and that of excitons. By comparing experimental data with our theoretical models we conclude that independent carriers dominate the dynamics of carrier escape in InAs/GaAs QD's.

2 Model

Let us consider a system of quantum dots in a semiconductor matrix (barrier) where electron/hole pairs are generated continuously mainly in the matrix at a pump rate P. Let n be the steady state concentration of electrons (holes) in the matrix. These carriers can undergo two possible processes: recombine at a rate of

$$W_{nr}^{(\mathrm{e,h})} = R' n^{(\mathrm{e,h})}, \tag{1}$$

or be captured by the quantum dots at a rate of

$$W_{c}^{(\mathrm{e,h})}(z) = R_c n^{(\mathrm{e,h})} D(z)[1 - f^{(\mathrm{e,h})}(z)]. \tag{2}$$

Here $R' = 10^9\ \mathrm{s}^{-1}$ is the recombination rate in the barriers, $R_c = 3 \times 10^{10}\ \mathrm{s}^{-1}$ is the capture rate, $f(z)$ is the occupation probability of the quantum dot states, and $D(z)$ is the normalized density of the quantum dot states. In the model we consider only the ground states of the quantum dots, since in our experiments we use relatively low excitation intensity, so that no photoluminescence was observed from the excited states. The ground-state energies have some distribution caused primarily by variations in the size of the quantum dots [14]. The distribution function for the ground-state energy levels of electrons and holes in the quantum dots is assumed to be Gaussian

$$D(z) = \frac{1}{\sqrt{2\pi}} \exp\left(-\frac{z^2}{2}\right). \tag{3}$$

We use renormalized energies, z, related to the hole energies $E^{(\mathrm{h})}$ via $E^{(\mathrm{h})} = E_m^{(\mathrm{h})} - zE_0^{(\mathrm{h})}$ and to the ground-state energies of the electrons $E^{(\mathrm{e})}$ in the quantum dots via $E^{(\mathrm{e})} = E_g - E_m^{(\mathrm{e})} + zE_0^{(\mathrm{e})}$. Here E_g is the bandgap of the barrier material, E_m is the energy difference between the maximum of the distribution of the ground-state energies and the band edge of the barrier, and $E_0^{(\mathrm{e})} = 21$ meV and $E_0^{(\mathrm{h})} = 10.5$ meV are the standard deviations of QD ground-state energies. The superscripts (e) and (h) refer to electrons and holes, respectively.

Carriers captured into the quantum dots can then undergo two further processes: they can either be thermally activated into the barrier layer at a rate of

$$W_{e}^{(\mathrm{e,h})}(z) = R_c N_0 D(z) f^{(\mathrm{e,h})}(z) \exp\left[\frac{-E_a^{(\mathrm{e,h})}(z)}{kT}\right], \tag{4}$$

or recombine radiatively at a rate of

$$W_r(z) = R N_D D(z) f^{(\mathrm{e})}(z) f^{(\mathrm{h})}(z), \tag{5}$$

Here $N_0 = 10^{14}\ \mathrm{cm}^{-2}$ is the density of GaAs barrier states, $N_D = 10^{10}\ \mathrm{cm}^{-2}$ is the areal density of QD's, E_a is the activation energy for thermal escape, and $R = 10^9\ \mathrm{s}^{-1}$ is the rate coefficient for radiative recombination in the quantum dots. The value of the activation energy, E_a, in Eq. (4) is equal to the difference between the ground-state energy in a quantum dot and the band edge energy of the barrier. It can be expressed in the form $E_a^{(\mathrm{e,h})} = E_m^{(\mathrm{e,h})} - zE_0^{(\mathrm{e,h})}$. In general, electrons and holes have different activation energies reflecting the asymmetry of the band offsets which automatically leads to different confinement energies, E_m.[1] Therefore as the escape rates are determined mainly by the confinement energies they will be different for electrons and holes and the assumption of equal escape rates used in most previous models is not justified.

The time evolution of the carrier concentrations in the barrier, n, and in the quantum dots, m, are described by the set of coupled rate equations [13]

$$\frac{dn^{(\mathrm{e,h})}}{dt} = P - W_{nr}^{(\mathrm{e,h})} - \int dz\, W_c^{(\mathrm{e,h})}(z) + \int dz\, W_e^{(\mathrm{e,h})}(z), \tag{6a}$$

$$\frac{dm^{(\mathrm{e,h})}(z)}{dt} = W_c^{(\mathrm{e,h})}(z) - W_e^{(\mathrm{e,h})}(z) - W_r(z). \tag{6b}$$

This is a system of two sets of nonlinear equations for electrons and holes. The rate equations for the electrons and holes cannot be solved independently since they are coupled via the radiative recombination terms W_r, which contains information on the occupation of the quantum dots by electrons and holes. In the case of the exciton character of the carrier dynamics, which is the second alternative in our theoretical consideration, the term responsible for the radiative recombination in Eq. (6b) has to be replaced by the exciton recombination rate $W_r(z) = RN_D D(z) f(z)$, where $f(z)$ refers to the distribution function of carrier (electron or hole) with the larger activation energy. Since our experiments were carried out under continuous excitation conditions, we restrict our theoretical analysis by considering only a steady state solution of Eqs. (6) which implies $dn/dt = 0$ and $dm/dt = 0$. The solution of Eqs. (6) is the population functions $f(z)$ for electrons and holes in the quantum dots for a given generation rate P.

In contrast to previous theoretical considerations, our model includes the following physical effects: (i) we clearly distinguish between the carrier relaxation in the form of independent electrons and holes or excitons; (ii) we consider the redistribution of charge carriers between the QD bound states via the GaAs barrier continuum states; (iii) the energy differences between the quantum dot states and the matrix (the confinement energies) are not used as fitting parameters but are calculated as a function of dot size [14]; (iv) when calculating the PL spectra we take into account the anticorrelation of the confinement energies of the electron and hole states caused by the variation of the dot size, i.e., smaller quantum dots have smaller confinement energies for both electron and hole states.

Though our approach is capable of calculating the temperature-dependent PL spectra [13], here we focus on the integrated PL intensity only, as it is the parameter which provides the most critical test of our two models. In the case of independent carriers the PL intensity is determined by

$$I_{\mathrm{ind}} \propto \int dz\, D(z) f^{(\mathrm{e})}(z) f^{(\mathrm{h})}(z), \tag{7}$$

where $f^{(\mathrm{e})}(z)$ and $f^{(\mathrm{h})}(z)$ are the occupation probabilities of the QD states for electrons and holes, respectively. Unlike the model of independent carriers, in the exciton model it is assumed that the electrons and holes are strongly bound and thus they always relax or are thermally released together. Under such circumstances the PL intensity is given by

$$I_{\mathrm{exc}} \propto \int dz\, D(z) f(z), \tag{8}$$

where $f(z)$ refers to the distribution function of carrier (electron or hole) with the larger activation energy. The remarkable difference between the exciton model and the model of independent carriers is that in the latter model the recombination rate depends on a probability of an electron or a hole being in a dot which is occupied by a carrier of the opposite charge, while the exciton model implies that the recombining partner is *always* present.

[1] The equality $E_m^{(\mathrm{e})} = E_m^{(\mathrm{h})} = 0.15$ eV in or case is occasional.

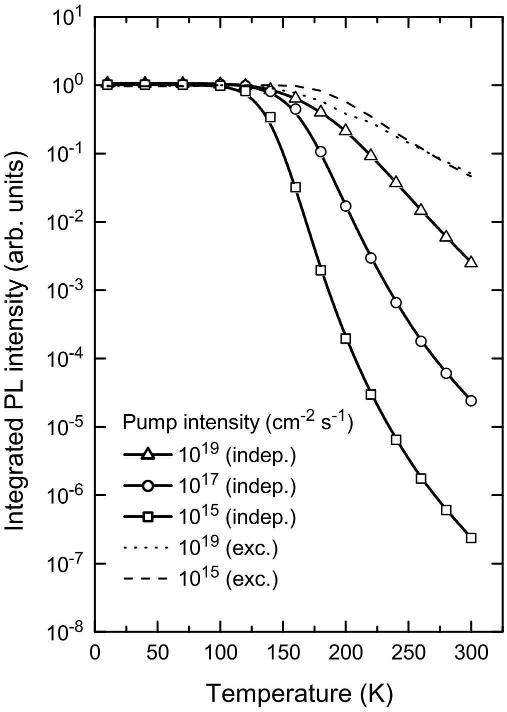

Fig. 1 Comparison between results of the independent-carrier model and those of the exciton model for temperature dependence of the integrated PL intensity at various pump intensities.

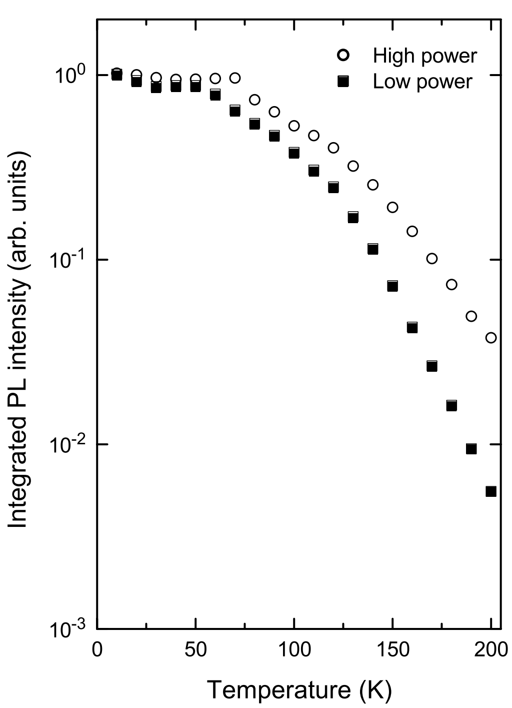

Fig. 2 Experimental data for temperature dependence of the integrated PL intensity taken from InAs/GaAs QD's using high and low excitation power densities (see text).

3 Results and discussion

We perform calculations of the temperature-dependent PL intensity taking the rate coefficients and material parameters relevant for relatively small QD's with the base size of approximately 10 nm (see Ref. [13] sample 617). The results of our calculations using the exciton model and the model of independent electrons and holes are shown in Fig. 1 for different values of the generation (pumping) rates. For both models the calculated PL intensities reproduce qualitatively the main features that are commonly observed, in particular, almost temperature-independent PL intensity at low T with subsequent quenching of the PL intensity with increasing temperature. The reasons for this general trend are obvious. At low temperatures thermal escape of charge carriers from the dots into the barriers does not occur sufficiently fast compared to the inverse radiative lifetime of carriers in the quantum dots. Hence the electrons and holes that are randomly captured by the quantum dots remain there until a carrier of the opposite charge appears in the same dot and recombination occurs. Therefore, radiative recombination from the quantum dots is the dominant recombination mechanism in this temperature range. With increasing temperature some carriers are thermally activated into the barrier states giving rise to either non-radiative or radiative recombination in the barriers. Despite the large activation energy (~ 150 meV), the thermal depopulation of the QD states already takes place at 100 K due to the high density of states in the barrier. This results in the reduction of the PL intensity for temperatures greater than 100 K.

Our aim is, however, to find out how crucial is the assumption on the independent carrier or exciton character of dynamics for the predicted efficiency of radiative recombination. The difference in the predictions of these two models becomes pronounced at higher temperatures and at low carrier density, when a significant fraction of the quantum dots are no longer occupied because of the thermal excitation into the barrier and subsequent non-radiative or radiative loss in the barriers. While the exciton model predicts the thermal quenching of PL intensity almost independent of the pump intensity (Fig. 1), the independent carrier model suggests rather strong dependence of the PL efficiency on the pump intensity at T above 100 K. In particular, reduction of the pump intensity by two orders of magnitude leads to the reduction of the room-temperature PL efficiency by two orders of magnitude (compare triangles and circles or circles and squares in Fig. 1). This observation is the most striking difference between the two models and it can be understood in the following way. In the exciton description the recombination partner for an electron/hole is always available, whereas in the case of separate carriers the probability of an electron/hole finding a recombination partner in the same dot decreases with increasing temperature due to the thermal depopulation of the QD states thus increasing the chance for non-radiative recombination. Therefore, depending on the excitation density, it is anticipated that at high temperatures the photoluminescence intensity predicted by the exciton model will be much larger than that calculated for the model of independent electrons and holes.

To judge which of these two models is correct we have compared our theoretical results with the measured thermal quenching of the PL intensity at different excitation intensities. We performed such measurements on buried InAs/GaAs QD's with a CW HeNe laser using the excitation power densities 0.01 and 160 W cm^{-2}. Results of the measurements are shown in Fig. 2. Experimental details can be found in Ref. [13]. The experimental data in Fig. 2 clearly favors the independent carrier model, which predicts that the thermal quenching of the PL intensity *depends* on the excitation power.

4 Conclusions

We have carried out experimental and theoretical studies of the temperature dependent optical properties of InAs/GaAs quantum dots paying particular attention to the nature of the carrier dynamics and relaxation processes. The optical measurements show distinct variations of the PL efficiency with excitation density that are best described by the theoretical model in which the carriers are treated as independent electrons and holes rather than correlated electron-hole pairs (excitons).

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