## Carrier statistics in quantum dot lasers

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The description of quantum dot ensembles using solely average carrier populations is insufficient. Bookkeeping of the probability of all micro-states and soluiton of the master equations for the transitions between them allows proper modeling of the phonon bottleneck and laser properties. We predict that single exciton shows gain and calculate the dependence of threshold current density of the type of capture process. The gain-current relation in quantum dot lasers is linear.

1. Quantum dots represent a unique electronic system which was recently successfully implemented for novel semiconductor lasers [1-4]. The charge carriers populate discrete electronic levels and (at least at sufficiently low temperature) all dots in an ensemble are laterally decoupled from each other. Therefore an event in a quantum dot, e.g. a recombination process, does not depend on the average carrier density (involving all other dots) but only on the particular population of the present dot with electrons and holes [5]. The ensemble has to be averaged over the probability distribution of the different micro-states. Different probability distributions of micro-states can have the same average carrier population but show different properties like recombination current. In order to illustrate this point with a simple example, we compare two QD ensembles with the same average carrier population: in ensemble I all electrons are in different dots than the holes: no radiative recombination takes place. In ensemble II electrons and holes populate the dots by pairs, leading to radiative recombination. We further discuss modeling of the phonon bottleneck effect.

**2**. Since the epitaxially created self-ordered QDs [6,7]are in the strong confinement regime, it is adequate to model electronic levels in the single particle picture, i.e. electrons and holes populate single particle levels. Ground state luminescence is said to originate from the radiative recombination of an electron and a hole in their respective single particle ground states (exciton). The lifetime of the exciton shall be denoted be  $\tau_X$ . The Coulomb correlation shall not fundamentally alter that picture. One effect is the shift of recombination energy of the biexciton (two electrons and two holes in their spin-degenerate ground states) with respect to that of the exction. For InAs/GaAs quantum dots this shift is expected to be small (< 2 meV) [8]; for II-VI compounds this shift will be larger. The radiative lifetime  $\tau_{XX}$  of the biexciton in the strong confinement limit is  $\tau_{XX} = \tau_X/2$  (as if two independent exciton decay); shortening of this time by Coulomb interaction is neglected here but could be included in our model.

A micro-state represents one particular population of the quantum dot levels with carriers, e.g. an empty dot (0, 0, ...), a dot with an electron-hole pair in the ground state (1, 0, ...), or a dot with an electron-hole pair in the first excited state (0, 1, ...). As visualized in Fig. 1, the latter micro-state can make two transitions: radiative decay  $(0, 1, ...) \rightarrow (0, 0, ...)$  and inter-sublevel scattering  $(0, 1, ...) \rightarrow (1, 0, ...)$ . The ensemble state is described by the probability  $p_n$  to find a micro-state *n* in the ensemble of all dots (the total number of dots shall be large),  $\sum_{\text{all }n} p_n = 1$ . The probability  $p_n$  can be also thought of as the time average of the micro-states one dot undergoes during a sufficiently long time interval, i.e. the quantum dot ensemble

is ergodic. **3.** The delayed energy relaxation in quantum dots, the so called "phonon bottleneck" effect has attracted much attention because of its potentially detrimental impact on performance of high speed devices. From time resolved experiments with resonant excitation of excited zero-dimenstional states of InAs/GaAs self ordered quantum dots [6.7] inter-

states of InAs/GaAs self-ordered quantum dots [6,7] intersublevel scattering times of 25–40 ps are found [9]. Ground state luminescence rise time is similarly fast (40 ps) for nonresonant excitation in the barrier. However, a modeling of such transients requires description with master equations for the micro-states [10]. If a conventional rate equation model (CRE) for the average population of sublevels is used in connection with a short inter-sublevel scattering time  $\tau_0$ , the transient of the excited state luminescence displays a fast and non-exponential decay. The experimental decay of luminescence from the first excited state is exponential as predicted by the master equations for the microstates (MEM) (Fig. 2).

The luminescence of the ground state is composed of photons from excitons and biexcitons. In Fig. 3 we decompose the calculated transient of an initially completely filled ground state into the excitonic and biexcitonic parts, which could be experimentally observed if both recombination energies are sufficiently separated. The biexciton transient is strictly exponential with a time constant  $\tau_{XX} = \tau_X/2$ . The excitonic luminescence increases first when excitons start to



**Figure 1.** Scheme of the transitions between micro-states.  $\tau$  is the lifetime for all radiative transitions.  $\tau_0$  is the inter-sublevel scattering time.



**Figure 2.** Experimental decay of the excited state luminescence from InAs/GaAs self-ordered quantum dots. Lines are fits with master equations for the micro-states (MEM) and conventional rate equations (CRE), both for an inter-sub-level scattering time  $\tau_0 = 30$  ps.



**Figure 3.** Calculated luminescence decay of completely filled ground state. Different lines are total intensity and intensity on exciton and biexciton recombination lines  $(I_{\text{total}} = I_X + I_{XX})$ .

be created by the  $XX \rightarrow X + \gamma$  process; eventually it decays exponentially with the time constant  $\tau_X$ .

**4**. The recombination current in a quantum dot ensemble cannot be described using solely the average electron and hole densities. Throughout the literature the bulk recombination rate [11] is used for the recombination current which is essentially a bimolecular expression

$$j = \frac{2eN_D}{\tau_X} f_e f_h, \tag{1}$$

having a maximum of  $j = 2eN_D/\tau_X$  for the fully occupied ground state.  $0 \le f_{e,h} \le 1$  denotes the average filling of

the ground state with electrons and holes. As already motivated in the introduction, the precise distribution of carriers over the dots has to be known to properly obtain the recombination current

$$j_{r} = \frac{N_{X}}{\tau_{X}} + \frac{N_{X-}}{\tau_{X-}} + \frac{N_{X+}}{\tau_{X*}} + \frac{N_{XX}}{\tau_{XX}}$$
$$= \frac{N_{X} + N_{X-} + N_{X+} + 2N_{XX}}{\tau_{X}}, \qquad (2)$$

where  $N_X$  denotes the number of dots filled with an exciton,  $N_{X-}$  and  $N_{X+}$  the number of dots with negatively or positively charged excitons and  $N_{XX}$  those with biexcitons. The right equality is valid in the strong confinement limit where  $\tau_X = \tau_{X-} = \tau_{X+} = 2\tau_{XX}$ . If excited states are populated with carriers additional terms enter Eq. (2). The recombination current is monomolecular as opposed to Eq. (1).

The gain g of a quantum dot ensemble depends linear on the carrier density [12]. The dependence of total ground state luminescence intensity and gain on an external injection current (Fig. 4) is also linear (until saturation dominates), while Eq. (1) predicts  $g \propto \sqrt{j}$  (dotted line in Fig. 5, a). In Fig. 4 we assumed for simplicity that no charged dots exist. If exciton and biexction recombination (and absorption) energy are sufficiently separated (compared to their homogeneous broadening and the inhomogeneous



**Figure 4.** Total, excitonic and biexitonic luminescence intensity and gain of the ground state versus external injection current. A capture time from the barrier  $\tau_c = \tau_X/100$  and a barrier recombination channel with  $\tau_b = \tau_X$  have been assumed.



**Figure 5.** *a*) Gain of a single layer quantum dot ensemble as a function of injection current for uncorrelated (e + h, solid line) and correlated (X, dashed line) capture of electron and holes. For comparison the result from conventional (Eq. (1)) mean field theory (MF, dotted line) is shown. The gain is given in units of the maximum gain. The inset shows the relation between gain and carrier density *N* in the dot ensemble, being identical and linear for all models. *b*) Threshold current for both capture models and conventional theory as a function of coverage for a typical dot ensemble and a total loss of  $\alpha_{tot} = 10 \text{ cm}^{-1}$  as a function of area coverage.

ensemble broadening), gain on the exciton and biexciton recombination energy have to be distinguished. As can be seen in Fig. 4, *a*, already the exciton offers gain. With increasing current *j* excitons are created  $\propto j$  and biexcitons  $\propto j^2$ . At a current of  $1e/\tau_X$  per dot the exciton luminescence reaches its maximum; it decreases for larger currents since most dots become filled with biexcitons.

Charged dots contribute gain at a lesser price in recombination current because the extra charge carrier does not radiatively decay but decreases absorption. Consequently the gain-current relation depends on whether or not charged dots exist in the ensemble. This depends on the capture mechanism as shown in Fig. 5. The limiting cases of completely correlated and uncorrelated electron hole capture are considered. In the first case excitons are captured and only neutral dots exist, causing the higher threshold current. In self-organized quantum dots capture occures directly from the barrier as well as via the two-dimensional wetting layer in which excitons can form before capture. If not pure exciton capture is present, we expect the capture of electrons and holes to be correlated because charged dots represent a more attractive site for the opposite charge.

A typical ensemble of self-organized InGaAs/GaAs quantum dots [1] was assumed with  $\tau = 1$  ns, inhomogeneous broadening  $\sigma_E = 20 \text{ meV}$ , a vertical optical confinement factor  $\Gamma_z = 0.7\%$  and a total loss (including the loss at mirrors) of  $\alpha_{tot} = 10 \text{ cm}^{-1}$ ;  $\zeta$  describes the area filling factor with dots. For small  $\zeta \leq 1.3\%$  the maximum gain is insufficient to evoke lasing on the ground state. The threshold (for ground state lasing) goes smoothly to infinity for  $\zeta \rightarrow 1.3\%$  because a recombination channel in the barrier had been included in the model (see caption of Fig. 3). For large coverage, when the threshold current density is close to the transparency current density, the two capture scenarios differ by almost a factor of two. For small coverage, when the gain has to be close to maximum, the difference becomes smaller. The conventional approach (Eq. (1)) yields a falsely small threshold in any case. We note that the presence of charge carriers at zero injection (due to doping) decreases the threshold [12,13].

**5**. Fundamental properties of quantum dot lasers are insufficiently described by using solely average carrier densities. A detailed analysis based on bookkeeping of the microstates is required and has been worked out by us. Time resolved experiments involving inter-sublevel scattering can be properly modeled. New properties of quantum dot lasers result, i.e. linear gain-current relation and excitonic gain. The threshold current density is higher for excitonic capture as compared to uncorrelated electron hole capture.

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