

Theory of random population for quantum dots

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Carrier capture and recombination in quantum dots are random processes. Conventional rate equation models do not take into account this property. Based on our theory of random population we predict recombination spectra, transients, and gain of quantum-dot ensembles. Even with infinitely fast interlevel energy relaxation excited levels become considerably populated. The impact of a slowdown of energy relaxation is modeled and criteria for a conclusive experimental observation of a finite interlevel-scattering time are given. [S0163-1829(97)06515-6]

I. INTRODUCTION

The luminescence from various quantum-dot systems has been studied in detail as a function of excitation density. Among these systems are localized excitons in growth interrupted quantum wells,¹ strain-induced quantum dots,² and self-organized quantum dots.^{3,4} A common feature observed is that with increasing excitation density excited states are populated and luminescence from these states is monitored. In the case of localized excitons in quantum wells, the exciton and biexciton population have been investigated,¹ while for other systems transitions between excited states of equal^{2,4} and different³ quantum numbers for electrons and holes were observed. Typically luminescence from excited states is observed before the ground-state luminescence is saturated.

It is a common belief that for efficient energy relaxation into the lowest available state there will be no luminescence from excited states as long as the ground state is not saturated. Consequently, observation of luminescence from excited states before the lower-energy transition is saturated is attributed^{2,4} to a slowdown of interlevel scattering, known as the “phonon bottleneck”⁵ effect. In the following we will first analyze a typical rate equation model and then present our theory of random population (RP), which considers carrier capture and recombination as random processes. We will argue that even for infinitely fast energy relaxation within each dot excited states become populated before the lower state is saturated.

Throughout this paper we will assume that the energy separation between dot states is large compared to $k_B T$, so that *thermal* population of excited states can be neglected (low-temperature limit).

II. RATE EQUATION MODELS

Conventional rate equation models rely on a mean-field theoretical scheme in the sense that they assume level populations averaged over the quantum-dot ensemble. Various rate equation models may be considered to describe capture and recombination in quantum dots, taking into account different capture and relaxation processes, e.g., Refs. 4 and 6. We discuss here the simple “trickle-down” model, which leads to the typical result of rate equation models, namely,

vanishing occupation of excited states for fast energy relaxation.

Two nondegenerate energy levels for *eh* pairs are assumed. The population of the levels shall be f_1 and f_2 , with $0 \leq f_i \leq 1$. *eh* pairs shall be captured with a generation rate (excitation) G into the upper level. The radiative lifetime τ_r of the *eh* pairs in the ground and the excited states is assumed to be identical. The relaxation from level 2 to 1 is governed by the intrinsic relaxation time τ_0 . The rate equation model is then for the stationary case

$$-\frac{f_2}{\tau_r} - \frac{f_2(1-f_1)}{\tau_0} + G = 0, \quad (1a)$$

$$-\frac{f_1}{\tau_r} + \frac{f_2(1-f_1)}{\tau_0} = 0. \quad (1b)$$

This model is a simplified two-level version of the model presented in Ref. 4 for five levels with degeneracies. It shall serve only as a typical example; additional complexity does not alter its principal results. An analytical expression can be obtained for the solution of Eqs. (1a) and (1b). In the limit $\tau_0 \rightarrow 0$ the solution for $G < 1/\tau_r$ is

$$f_1 = G\tau_r, \quad f_2 = 0. \quad (2a)$$

In the case $1/\tau_r < G < 2/\tau_r$, the solution is

$$f_1 = 1, \quad f_2 = G\tau_r - 1. \quad (2b)$$

We note that for infinitely fast energy relaxation (or interlevel scattering) rate equation models generally yield vanishing population of excited levels as long as a lower level is not completely filled.

III. RANDOM POPULATION

The “trickle-down” rate equation model and other rate equation models that include additional capture mechanisms result in zero population of excited levels for infinitely fast energy relaxation ($\tau_0 \rightarrow 0$). Infinitely fast energy relaxation means that when a dot is filled with n electron-hole pairs, they are in the lowest n possible energy states at all times. However, such kinds of rate equation models, working with level populations averaged over the ensemble, do not describe a quantum-dot ensemble population appropriately.

Let us look at a simple example and assume that the external excitation is low, $G < 1/\tau_r$. The (large) total number of quantum dots shall be N_D . Thus $f_1 N_D$ quantum dots are filled with one electron-hole pair, the rest are empty. If now an additional eh pair is captured from the reservoir by the quantum-dot ensemble, two things can happen: If it is captured by an empty dot, f_2 remains zero. However, there is a finite chance that it is captured into one of the $f_1 N_D$ dots that are already filled with one eh pair, making $f_2 > 0$. This is a contradiction to the prediction of the rate equation model.

In the following we develop a theory based on the idea that the carrier capture by and recombination in quantum dots are essentially random processes. The RP model is based on the following assumptions: (i) We consider an ensemble of N_D dots. Each of the dots has M levels for electrons and holes. Each level is counted separately, even if some of them are degenerate in energy. (ii) Radiative transitions occur only between electrons and holes with the same level numbers and result in photons of energy E_n . (iii) The radiative lifetime τ_r for all radiative transitions E_n is the same. Nonradiative channels do not exist. (iv) The external excitation fills a reservoir (wetting layer, barrier) with a generation rate G . From this reservoir carriers are captured into the dots. Additionally carriers in the reservoir can recombine with a radiative lifetime τ_b . (v) All dots are neutral, i.e., only eh pairs are captured. Further below we will also consider separate capture of electrons and holes with identical capture times. (vi) The number of eh pairs in the reservoir shall be N_R . The time it takes to transfer one eh pair from the reservoir into one empty dot (to any of its M levels) is τ_c^0/N_R . Later we will use the term $\tau_c = \tau_c^0/N_R$. When the dot is completely filled with M eh pairs it cannot capture additional carriers. We assume a linear decrease of capture time with the dot filling; i.e., when the dot is populated with n eh pairs, the capture time is $(1 - n/M)^{-1} \tau_c^0/N_R$. We will also consider a model with constant capture time, independent of n . (vii) Interlevel energy relaxation is infinitely fast, i.e., when the dot is populated with n eh pairs, the lowest n levels for electrons and holes are filled. Further below we will consider finite energy relaxation times. (viii) The low-temperature limit applies; i.e., no carriers are reemitted by the dots into the reservoir and no excited states are thermally occupied. Depopulation of dots by the Auger effect is neglected and subject to further studies.

With the above assumptions the steady-state solution of the model can be given analytically. Conditions (i), (ii), and (iii) are valid for uncoupled dots in the strong confinement regime. Nonradiative recombination can be easily accounted for by interpreting τ_r and τ_b as effective lifetimes. Fast energy relaxation (vii) gives the *lower limit* of population of excited levels. In a numerical treatment no restrictions apply and different radiative lifetimes for the different transitions (iii), partly forbidden transitions (ii) could be taken into account. Capture of single carriers (v) and finite interlevel scattering time will be considered further below.

Typical realistic time constants are $\tau_r \approx \tau_b \approx 1$ ns and a capture time τ_c in the 10-ps range, $\tau_c \ll \tau_r$. For our numerical examples in the following we will use $\tau_b = \tau_r$ and $\tau_c = \tau_r/100$.

The ensemble is described with microstates. The number of quantum dots filled with n eh pairs shall be N_n^M . Thus

$$\sum_{n=0}^M N_n^M = N_D. \quad (3)$$

The probability of finding a dot with n eh pairs is $w_n^M = N_n^M/N_D$. The number N_{eh} of eh pairs in the entire ensemble, and the average population $\langle n \rangle$ are

$$N_{eh} = \sum_{n=0}^M n N_n^M, \quad \langle n \rangle = \frac{N_{eh}}{N_D} = \sum_{n=0}^M n w_n^M. \quad (4)$$

The recombination rate from the i th level ($i > 0$, empty dots do not contribute to the spectrum) is

$$R_i^M = \frac{1}{\tau_r} \sum_{m=i}^M N_m^M = \frac{N_D}{\tau_r} \sum_{m=i}^M w_m^M. \quad (5)$$

The total recombination rate R_D from the quantum dots is ($n > 0$)

$$R_D = \sum_{i=1}^M R_i^M = \frac{N_{eh}}{\tau_r} = \frac{N_D}{\tau_r} \langle n \rangle. \quad (6)$$

The recombination rate (spectrum) at a particular energy is

$$I_0^M(E) = \sum_{i=1}^M R_i^M \delta(E - E_i). \quad (7)$$

In this formula the actual energy degeneracies of the levels play a role. In order to fit the model to real quantum-dot ensembles with size fluctuations a Gaussian inhomogeneous broadening is introduced. We assume that variation of transition energies E_i with population is small compared to the inhomogeneous broadening, as found in recent experimental investigations.^{2,3} The spectrum is then given by

$$I^M(E) = \sum_{i=1}^M R_i^M G(E - E_i, \sigma_i), \quad (8)$$

where $G(E, \sigma) = (1/\sqrt{2\pi}\sigma) \exp(-E^2/2\sigma^2)$. We note that (since we work in the low-temperature limit) dots of different ground-state energy have the same population. This means that the carrier distribution is *nonthermal* due to the lack of interdot coupling. Such nonthermal carrier distribution has recently been found in quantum-dot lasers.⁷

The master equation for the N_n^M dots filled with n eh pairs in the random population model is ($0 < n < M$)

$$\begin{aligned} \frac{dN_n^M}{dt} &= \frac{(n+1)N_{n+1}^M}{\tau_r} - \frac{nN_n^M}{\tau_r} + \frac{N_R N_{n-1}^M}{\tau_c^0} \left(1 - \frac{n-1}{M} \right) \\ &\quad - \frac{N_R N_n^M}{\tau_c^0} \left(1 - \frac{n}{M} \right) = 0. \end{aligned} \quad (9a)$$

For $n=0$ and $n=M$ we have the special cases

$$\frac{dN_0^M}{dt} = \frac{N_1^M}{\tau_r} - \frac{N_R N_0^M}{\tau_c^0} = 0, \quad (9b)$$

$$\frac{dN_M^M}{dt} = -\frac{MN_M^M}{\tau_r} + \frac{N_R N_{M-1}^M}{\tau_c^0} \left(1 - \frac{M-1}{M}\right) = 0.$$

For the reservoir we have the additional condition

$$\frac{dN_R}{dt} = G - \frac{N_R}{\tau_b} - \frac{N_R}{\tau_c^0} \sum_{n=0}^M N_n^M \left(1 - \frac{n}{M}\right) = 0. \quad (10)$$

By iteratively solving the equation system for N_n^M we find

$$N_n^M = N_0^M \frac{1}{n!} \left(\frac{N_R}{\tau_c^0} \tau_r\right)^{n-1} \prod_{m=0}^{n-1} \left(1 - \frac{m}{M}\right). \quad (11)$$

From the normalization condition (3) and Eq. (10) N_R is determined:

$$\begin{aligned} \frac{N_R}{N_D} &= -\frac{M}{2} \frac{\tau_b + \tau_c}{\tau_r} + \frac{1}{2} \frac{G \tau_b}{N_D} \\ &+ \left(M \frac{G \tau_b}{N_D} \frac{\tau_c}{\tau_r} + \left[\frac{M}{2} \frac{\tau_b + \tau_c}{\tau_r} - \frac{1}{2} \frac{G \tau_b}{N_D}\right]^2\right)^{1/2}. \end{aligned} \quad (12)$$

Subsequently all N_n^M can be calculated.

If in another model of carrier capture it is assumed that the capture time is independent of n , the solution is given by

$$\frac{N_R}{N_D} = \frac{G \tau_c}{1 + \tau_c/\tau_b} \approx G \tau_c. \quad (13)$$

In the limit $M \rightarrow \infty$ the capture probability also does not depend on the filling and the $(1-n/M)$ terms are identical to 1. Then the same formula (13) applies. Furthermore, the probability to find a quantum dot with n *eh* pairs is then given by a Poisson distribution

$$w_n^M = \frac{\lambda^n}{n!} \exp(-\lambda), \quad \text{with } \lambda = \langle n \rangle = \frac{G \tau_r}{N_D} \frac{1}{1 + \tau_c/\tau_b} \approx \frac{G \tau_r}{N_D}. \quad (14)$$

For finite M we find in the low excitation limit $G \rightarrow 0$,

$$N_R \rightarrow \frac{G \tau_b}{1 + \tau_b/\tau_c} \approx G \tau_c, \quad (15)$$

since usually the capture time is much faster than the recombination time in the barrier. In the high excitation limit, $G \rightarrow \infty$, when for finite M all quantum dots are saturated, we find $N_R \rightarrow G \tau_b$.

IV. RECOMBINATION SPECTRUM

The recombination spectrum for a particular excitation G is obtained from Eq. (8). For simplicity we assume the quantum dots to be disklike, so that their energy levels can be well described by a two-dimensional harmonic oscillator model. The K energy states E_k are $g_k = 2(k+1)$ degenerate. The total number of levels is $M = K(K+1)$

$$E_k = E_h + (k+1)\hbar\omega, \quad k = 0, 1, \dots, K-1. \quad (16)$$

The energetic broadening σ_k depends on the variation σ_h of the offset energy E_h , e.g., due to disk thickness variation, and the fluctuation σ_ω of $\hbar\omega$, e.g., due to disk radius fluctuations

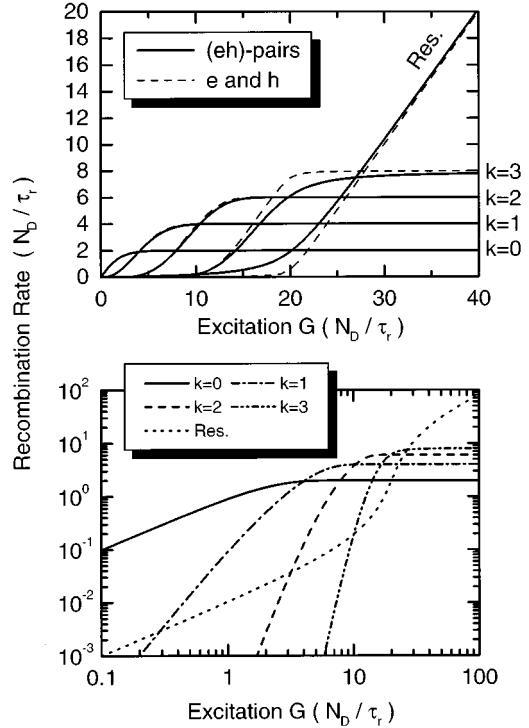


FIG. 1. Intensity (recombination rate) on the dot states E_k , $k=0, \dots, 3$ and the reservoir (Res.) as a function of external excitation density in linear (a) and double logarithmic (b) plots. Solid (dashed) curves in (a) are for simultaneous (separate) electron and hole capture.

$$\sigma_k = \sqrt{\sigma_h^2 + (k+1)\sigma_\omega^2}. \quad (17)$$

For our numerical example we use $\sigma = 20$ meV for all transitions, and $\hbar\omega = 60$ meV. In Fig. 1 we show the recombination rates for different recombination energies E_k and for the reservoir as a function of excitation G . For low excitation ($G \ll N_D/\tau_r$) only luminescence from the ground state is present. Due to the random nature of capture and recombination the intensity of an excited level E_{k+1} rises well before the intensity of the energetically lower state E_k saturates, even for the assumed infinitely fast energy relaxation within the dots. However, the E_{k+2} level does not start to exhibit significant intensity before E_k saturates. In Fig. 2 spectra are shown for quantum dots with $K=5$ at different excitation levels for RP theory and the “trickle-down” rate equation. Thus in the light of this model, only the definite observation of luminescence from the E_{k+2} level before E_k is saturated provides a fingerprint of finite energy relaxation time. The conclusion on a “phonon-bottleneck” effect from the observation of luminescence from the E_{k+1} state before the lower transition is saturated⁴ is not valid.

V. LASER PROPERTIES

From the population statistic the gain spectrum can be calculated. Here, we consider the gain of the ground-state transition (the two lowest levels). The gain of the quantum dot ensemble is given by

$$g = C_g(2f-1), \quad (18)$$

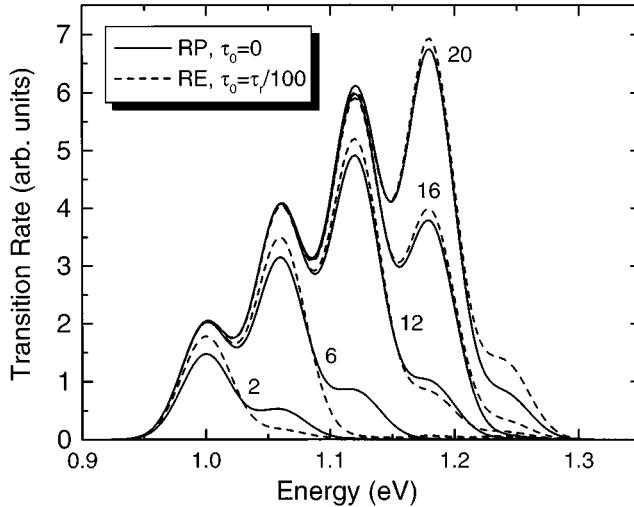


FIG. 2. Spectra for the level populations of Fig. 1 for $G\tau_r/N_D = 2, 6, 10, 16$, and 20 for RP theory (solid lines) and rate equation model for five states and $\tau_0/\tau_r/100$ (dashed line).

where C_g is a constant collecting all prefactors, and f is the population probability of the ground state. In terms of our model,

$$f = \frac{w_1^M}{2} + \sum_{n=2}^M w_n^M = 1 - \frac{w_1^M}{2} - w_0^M. \quad (19)$$

Therefore

$$g = C_g (1 - w_1^M - 2w_0^M). \quad (20)$$

For $M \rightarrow \infty$ the excitation dependence of gain is

$$\begin{aligned} g &= C_g \left\{ 1 - \left[2 + \frac{G\tau_r}{N_D} \right] \exp\left(-\frac{G\tau_r}{N_D}\right) \right\} \\ &\approx C_g \{ 1 - [2 + \langle n \rangle] \exp(-\langle n \rangle) \}. \end{aligned} \quad (21)$$

In Fig. 3 we compare the gain versus excitation curves for quantum dots with different numbers of levels. Already $M=20$, i.e., $K=4$, for the two-dimensional harmonic oscillator, is very close to the limit $M \rightarrow \infty$. As the solid curve we have included the gain obtained from the conventional rate equation model⁴ for $\tau_0=0$, which fills all dots equally from the bottom. In RP theory the gain of the ground state at an injection current $2N_D/\tau_r$ is $C_g[1-4\exp(-2)] \approx 0.46C_g$, i.e., only about half the saturated value, for $M \rightarrow \infty$. The solid circles denote the gain from the “trickle-down” rate equation model⁴ with $K=5$ and $\tau_0=\tau_r/100$. Obviously this model overestimates the gain and underestimates the current for gain saturation.

The transparency current I_{tr} , for which $g=0$, has a similar value for all models,

$$I_{tr} \approx eN_D/\tau_r. \quad (22)$$

We note that this value is two times larger than the usual result from mean-field theory, where transparency is obtained for electron and hole population probabilities $f_n = 1 - f_v = 1/2$, with an associated recombination current

$$I = 2eN_D/\tau_r f_v (1 - f_v) = \frac{1}{2} eN_D/\tau_r.$$

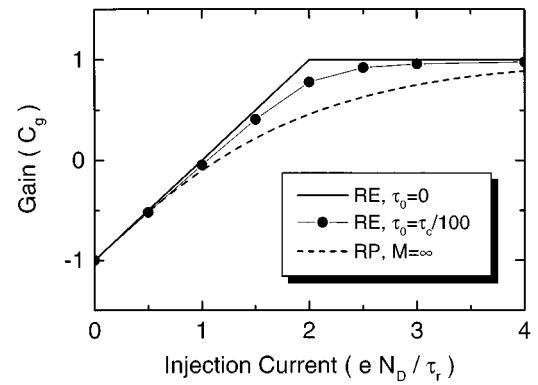


FIG. 3. Gain on the ground state (two levels) for rate equation models with infinitely fast energy relaxation (solid line), “trickle-down” rate equation model with five states (Ref. 4) for $\tau_0 = \tau_r/100$, and random population theory for $M = \infty$.

VI. SEPARATE CAPTURE

In this section we discuss separate capture of electrons and holes. We assume identical capture times for electrons and holes, which leads to charge neutrality of both the reservoir and the dot ensemble. $N_{n,m}^M$ shall be the number of dots (n,m) being populated with n electrons and m holes; for $n \neq m$ the dot is charged. We immediately find $N_{n,m}^M = N_{m,n}^M$. The master equations (9) can be easily modified for the separate capture of electrons and holes. We note that the recombination rate from the (n,m) dot is $\min(n,m)N_{n,m}^M/\tau_r$. For the resulting equation system a simple analytical solution cannot be given. In Fig. 1(a) we compare the numerical solution for $K=4$ with the model of eh pair capture. Only for the highest state and high excitation the two models exhibit a significant difference: the highest state saturates quicker and the intensity from the reservoir only starts at higher excitation density. In the following sections we will therefore use the model of eh -pair capture.

Although the luminescence spectra do not differ significantly for the two capture models, we mention a peculiarity of the population of charged dots. In Fig. 4 we depict the population probabilities $w_{n,m}^M = N_{n,m}^M/N_D$ and the number of carriers in the reservoir for dots with two levels ($M=2$) as a function of excitation. At low excitation $G \rightarrow 0$ simply charged dots (N_{10}^2) are as frequent as empty dots (N_{00}^2); additionally double charged dots are present (N_{20}^2). For $G \equiv 0$, however, the solution is of course $N_{00} = N_D$. This is not a true discontinuity of the solution since for $G \rightarrow 0$ it takes longer and longer time to “load” the charged dots with carriers and reach the steady state. The transparency current in this case is $I_{tr} = \frac{5}{8}eN_D/\tau_r$, i.e., almost a factor of 2 smaller than for an ensemble of neutral dots and slightly larger than the result from mean-field theory.

VII. FINITE INTERLEVEL SCATTERING

Nonzero interlevel scattering times can be included in the model numerically using Monte Carlo simulations of capture, scattering, and recombination. We assume that any scattering event between a filled upper level and an empty lower level is described by the same scattering time τ_0 . We note that the probability w_n^M to find a dot with $n eh$ pairs does not

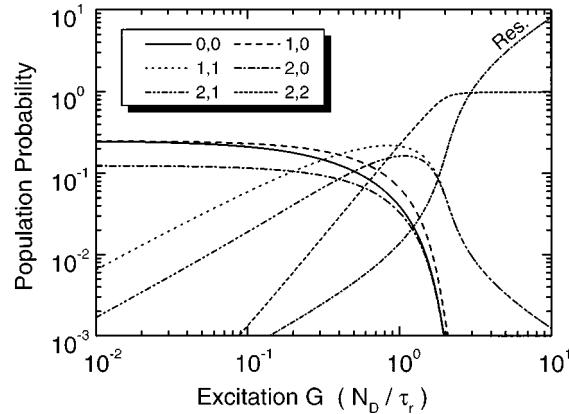


FIG. 4. Probability to find \$(n,m)\$ dots with \$n\$ electrons and \$m\$ holes as a function of excitation for \$M=2\$ for separate electron and hole capture. Curve labeled Res. describes the number of carriers \$N_R/N_D\$ in the reservoir.

depend on \$\tau_0\$ and remains unchanged from the solution given in Eq. (11). The effect of nonzero \$\tau_0\$ is that the \$n\$ \$eh\$ pairs are no longer in the \$n\$ lowest levels; actually there are now \$p_n^M = \binom{n}{M} = n!/M!(n-M)!\$ possible distributions of the \$n\$ \$eh\$ pairs over the \$M\$ available levels. In the limit of very slow interlevel energy relaxation, \$\tau_0 \rightarrow \infty\$ and \$\tau_0/\tau_r \gg 1\$, the \$eh\$ pairs are equally distributed among all levels and the probability of finding a dot in any of the \$p_n^M\$ possible states is identical and given by \$w_n^M/p_n^M\$.

The impact of the interlevel-scattering time on the luminescence from an excited state is first shown for the simplest example, a dot with two states \$K=2\$. In Fig. 5 we depict the ratio \$I_0/I_1\$ of the recombination rates from \$E_0\$ (ground state) and \$E_1\$ (excited state) as a function of \$\tau_0\$ for two different excitations. For fast relaxation \$\tau_0 \rightarrow 0\$ the ratio tends towards the limit obtained analytically from Eq. (11). For slow relaxation \$\tau_0 \rightarrow \infty\$ the ratio reaches \$1/2 = g_0/g_1\$, determined by the state degeneracies.

A comparison of the recombination rates for zero and finite scattering time for the \$K=4\$ dot is visualized in Fig. 6. For finite interlevel relaxation the saturation of lower levels is slower and excited states gain intensity at lower excitation

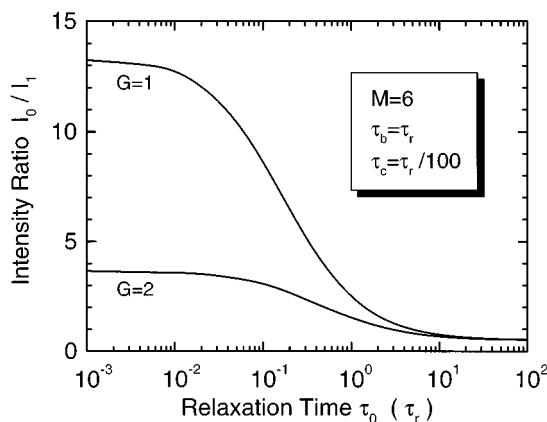


FIG. 5. Ratio of the luminescence intensity on the \$k=1\$ and \$k=0\$ states for a quantum dot with \$K=2\$ states (\$M=6\$ levels) for two different excitation densities (given in units of \$N_D/\tau_r\$) as a function of the interlevel-scattering time \$\tau_0\$.

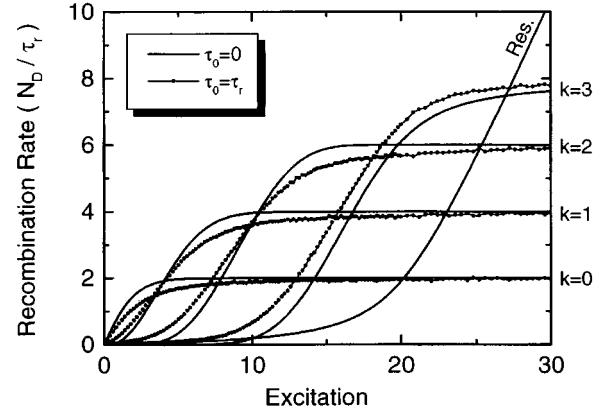


FIG. 6. Intensity of luminescence from quantum-dot states with \$K=4\$ states as a function of excitation for different interlevel scattering times (\$\tau_0=0\$, \$\tau_0=\tau_r\$). For \$\tau_0=0\$ the curves are identical with those from Fig. 1(a). The scatter in the points for \$\tau_0 \neq 0\$ is due to the Monte Carlo method used.

density. The \$E_{k+2}\$ level now appears before the \$E_k\$ level is saturated as a fingerprint of the slowdown of relaxation. Spectra for different excitations and \$\tau_0=0\$ and \$\tau_r\$ are visualized in Fig. 7. We note that a modified model, allowing capture into the top quantum-dot level only and subsequent interlevel relaxation, yields almost identical spectra for the parameters used in Fig. 7.

VIII. RECOMBINATION DYNAMICS

The recombination dynamics can also be modeled with RP theory by setting \$G=0\$ in Eq. (10) and solving the time dependence (9a), (9b), and (10) numerically for \$\tau_0=0\$ using a Runge-Kutta algorithm. Let us assume that the excitation \$G\$ is terminated at the time \$t=0\$, when the dot ensemble is characterized by the steady-state carrier distribution \$N_n^M(0)\$

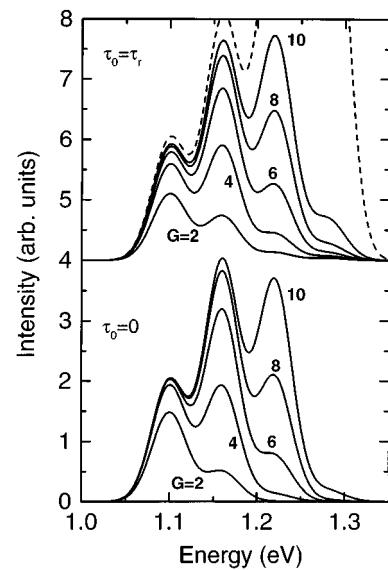


FIG. 7. Spectra for quantum-dot with \$K=4\$ states for different excitations (\$G=2, 4, 6, 8\$, and \$10 N_D/\tau_r\$) and interlevel-scattering times. The dashed line represents the luminescence spectrum from a completely saturated dot ensemble.

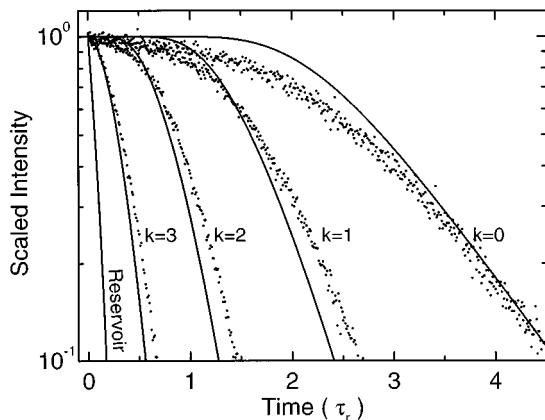


FIG. 8. Transients for $M = 20$ ($K = 4$) dot in RP theory; steady-state excitation for $t < 0$ is $G = 20 N_D / \tau_r$. The interlevel scattering times are $\tau_0 = 0$ (solid line), and $\tau_0 = \tau_r$ (points). The scatter in the points for $\tau_0 \neq 0$ is due to the Monte Carlo method used.

and $N_R(0)$. In Fig. 8 the time development of the $K = 4$ quantum dot is shown for a steady-state initial population due to $G = 20 N_D / \tau_r$. For infinitely fast interlevel scattering lower levels remain at their steady-state intensity as long as the closest excited state did not significantly decay. For non-zero interlevel energy relaxation time, lower levels start to decay immediately due to the partly suppressed refilling of lower levels emptied by radiative recombination. Transients

showing such an effect for the first excited and ground quantum-dot state were reported in Ref. 8.

IX. CONCLUSION

We have developed a theory of population of quantum-dot levels based on the random nature of capture, interlevel energy relaxation and recombination processes. The level populations obtained from models with simultaneous and separate capture of electrons and holes are very similar. We find that still under the assumption of infinitely fast energy relaxation within each dot, the next excited state is already significantly populated before the energetically lower state saturates. This behavior is in contrast to the results of conventional rate equation models. For nonzero interlevel-scattering time the saturation of levels becomes slower and higher excited states become populated earlier.

In the light of our model we conclude that a clear spectroscopic manifestation of the “phonon-bottleneck” effect (slow energy relaxation) is present only if (1) the luminescence from the E_{k+2} state (or higher states) is observed before E_k is saturated. (2) In the transient from the steady state (all) lower levels start to decay right after termination of the excitation.

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