Carrier relaxation dynamics in quantum dots: Scattering mechanisms and state-filling effects

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Stressor-induced $In_xGa_{1-x}As$ quantum dot structures of high structural quality allow a detailed experimental investigation of carrier relaxation between distinct zero-dimensional quantized states. Time-resolved photoluminescence studies combined with appropriate model calculations show that state filling effects, Coulomb scattering, and acoustic phonon scattering determine the relaxation scenario in a way characteristic for a zero-dimensional electronic system. These investigations allow a quantitative estimation of the inter-dot-level relaxation rates mediated by (i) Coulomb scattering and (ii) acoustic phonon scattering. [S0163-1829(97)08907-8]

The role of system dimensionality in the thermalization process of photoexcitations has been a topic of intense research efforts for many semiconductor systems.[1] The relaxation dynamics in a zero-dimensional (0D) quantum dot (QD) semiconductor system is expected to qualitatively differ from higher-dimensional systems, since the density-of-states (DOS) is a series of δ functions. This discrete DOS leads to severe energy conservation problems for scattering of 0D electrons with optical phonons and also with other 0D electrons.[2–5] In addition, the finite degeneracy of each QD state leads to state filling effects already when few carriers populate the lowest dot states.[6,7] Thus carrier relaxation from excited dot states to energetically lower dot states (inter-dot-level relaxation) should be hindered already at low excitation densities (Pauli blocking).

During recent years several groups have performed time-resolved optical experiments on various QD structures to study the relaxation and recombination dynamics of carriers confined in three dimensions.[8–16] However, the carrier relaxation scenario into and within the QD potential is still discussed controversially. In addition, for semiconductor QD's a systematic comparison between experimentally obtained time-resolved optical data and calculations or a modeling of the carrier relaxation scenario has not been done so far to our knowledge.

Lipsanen, Sopanen, and Ahopelto[6] reported on the fabrication of strain-induced $In_xGa_{1-x}As$ QD's with remarkably high quantization but low inhomogeneous broadening effects. As discussed in the present paper, these strain-induced $In_xGa_{1-x}As$ QD structures offer the possibility to carry out experimental studies of the relaxation scenario of photocreated carriers into and within zero-dimensional semiconductor states by performing time-resolved photoluminescence (PL) experiments. We can model the characteristic PL transients of the individual dot transitions by solving a simplified rate equation system. It is shown that Pauli blocking induced by few carriers and 0D-2D Coulomb scattering are crucial ingredients to understand the temporal evolutions of the distinct QD PL transitions. Quantitative estimates of acoustic-phonon scattering rates and Coulomb scattering rates being responsible for the inter-dot-level relaxation are given.

The investigated QD sample has the following structure.[18] A single 7-nm-thick $In_{0.1}Ga_{0.9}As$ layer is embedded within GaAs. The GaAs top barrier has a thickness of 5 nm. InP islands were grown *in situ* on this top layer. These islands, which act as stressors, are $\approx$75 nm wide and $\approx$22 nm high as determined by atomic force microscopy. The island density is approximately 2×10^9 cm^{-2}. The inset of Fig. 1 shows the strain-induced, paraboliclike lateral potential profile for electrons in the conduction band and induced in the $In_xGa_{1-x}As$ quantum well (QW) below the InP stressors. The calculated energetic difference between the lowest confined QD state and the first quantized state in the QW is 64

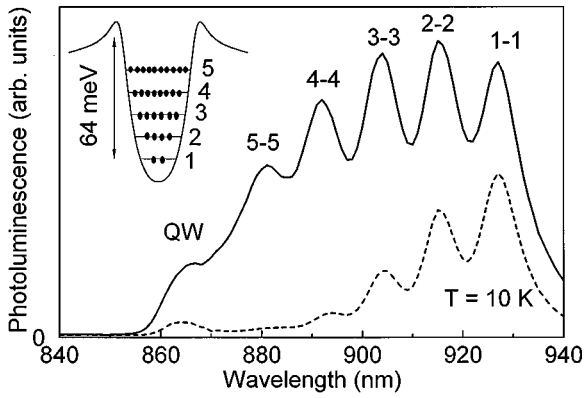

FIG. 1. Low-temperature photoluminescence spectra of the $In_xGa_{1-x}As$ quantum dot sample for low (dashed line) and high (solid line) excitation intensities. The inset shows the strain-induced lateral potential for electrons in the conduction band. The number of filled circles on each quantum dot level i gives the degeneracy $D_i=2i$.

meV. Each quantized dot level is characterized by a quantum number for the angular momentum ($m=0, \pm 1, \pm 2, \ldots$ or $\Sigma,\Pi,\Delta,\ldots$) and a radial quantum number ($n=1,2,3,\ldots$).[17] As shown in Ref. 17 some of these levels are degenerate. In the following we only distinguish between nondegenerate QD levels as indicated in the inset of Fig. 1 ($i=1,2,3,\ldots$). The number of filled circles drawn on each QD level indicates the respective degeneracy $D_i=2i$ taking into account spin degeneracy.

The low-temperature PL spectra of the QD sample taken at an excitation wavelength of 805 nm and excitation intensities of 50 W/cm^2 (dashed curve) and 250 W/cm^2 (solid curve) are shown in Fig. 1. First of all the PL line at ≈ 865 nm can be attributed to exciton recombination from the $In_xGa_{1-x}As$ QW outside the region covered by the InP islands. As already shown by Lipsanen, Sopanen, and Ahopelto[6] and Tulkki and Heinämäki[17] the five spectrally well-separated PL lines seen in the high-intensity spectrum at longer wavelengths are due to electron-hole recombination between distinct QD states of the conduction and valence band. As shown in Ref. 17 a conservation rule $\Delta i=0$ holds to a good approximation for the interband QD transitions. Accordingly, we have labeled the QD PL transitions in Fig. 1 by i-i. From the energetic spacing between the two energetically lowest QD PL lines we calculate a radial quantization energy within the lateral parabolic potential of $\hbar\omega_e=11$ meV for electrons in the conduction band.

We have also performed absorption and photoluminescence excitation (PLE) experiments (not shown here) detecting the PL from the 1-1 transition.[12] For photon energies below the onset of the QW absorption, i.e., in the spectral region of the QD transitions, we do not find detectable absorption. This is probably due to the fact that even in the absence of light the QD states in the valence band are at least partially occupied by holes bleaching the interband QD transitions. These holes are a consequence of the unintentional p doping of the GaAs buffer. If this assumption is correct, the PL dynamics should be mainly determined by the relaxation of electrons in the conduction-band.

In order to gain insight into the relaxation and recombination scenario we have performed time-resolved low-temperature PL experiments using a 76-MHz Kerr-lens mode-locked Ti:sapphire laser producing 110-fs laser pulses focused to a spot approximately 100 μm in diameter. The central photon wavelength of the excitation laser is 800 nm. We have adjusted the excitation intensity to get a PL spectrum comparable to the high-intensity spectrum shown in Fig. 1. As shown in the following, this allows the investigation of the carrier relaxation dynamics between distinct QD states starting with several fully occupied QD levels. The PL transients at selected wavelengths are recorded by using a monochromator and a streak camera providing a temporal resolution of ≈ 13 ps. In Fig. 2(a) the results of time-resolved PL measurements are shown for the QW PL and the four lowest QD transitions (1-1, 2-2, 3-3, and 4-4) in a semilogarithmic plot.

We first give a qualitative discussion of the time-resolved PL spectra of Fig. 2(a). The rise times of all recorded PL transients are limited by the temporal resolution of the detection system; this shows that carriers relax very efficiently even to the lowest QD states. In accordance with Ref. 4 we

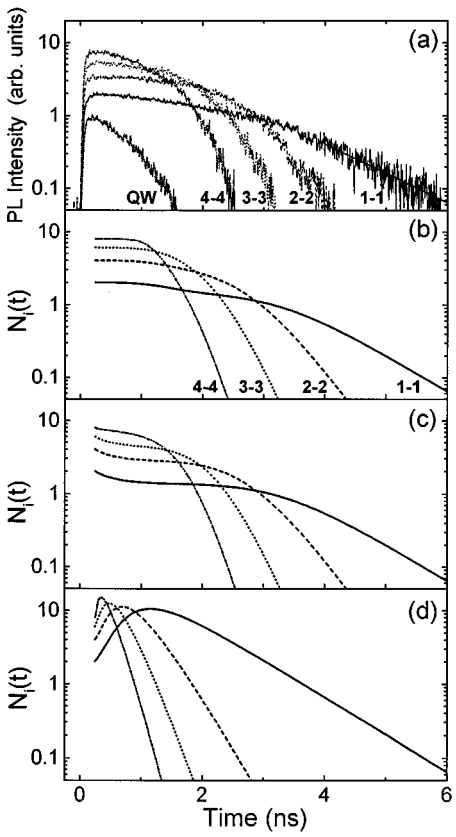

FIG. 2. (a) Time-resolved photoluminescence spectra of the four lowest QD transitions and of the uncovered $In_xGa_{1-x}As$ quantum well for a high excitation condition comparable to the case shown as the solid line in Fig. 1. (b) Calculated temporal evolution of the mean number of electrons $N_i(t)$ occupying the four lowest quantum dot levels choosing $\tau_{ph}=570$ ps and $\tau_{0D\text{-}2D}=1$ ps at $t_0=0.25$ ns as parameters for acoustic phonon and Coulomb scattering, respectively. Details of the model calculations are described in the text. (c) Same calculations as under (b) only neglecting Coulomb scattering ($R=0$). (d) Same calculations as under (c) only neglecting state filling effects ($f\equiv 1$).

assume that Coulomb scattering between carriers confined in 0D QD states and carriers located in the 2D QW states is responsible for the fast relaxation down to the lowest QD states. We assume for these high excitation densities that after this relaxation the lowest QD level ($i=1$) is occupied by two electrons. Accordingly, in Fig. 2(a) the y scale is chosen in such a way that the PL intensity of the 1-1 transition is equal to 2 right after the signal rise. The absolute PL intensities of the higher-energetic transitions (2-2, 3-3, and 4-4) then reach values close to the respective values of the calculated degeneracies $D_i=2i$. This experimental result means that the lowest QD levels are almost fully occupied at the maxima of the PL transients. After the signal rise the QD PL transients show a plateaulike behavior before they successively start to decay after 1.5–3.5 ns. The temporal duration of these ''plateaus'' can be shortened by decreasing the excitation intensity (not shown here). The following model calculations of the carrier relaxation show that this plateaulike behavior can be explained by properly taking into account state filling of the respective QD levels and also 0D-2D Coulomb scattering. Figure 2(a) further shows that

after ≈ 4 ns there is only PL from the lowest QD transition 1-1 left. Since at these late times feeding processes from higher states have ceased, the 1-1 PL decays exponentially. We thus identify the respective decay time of $\approx 860 \pm 25$ ps as the pure recombination lifetime τ_{rec} of the lowest QD transition.[19] Due to the facts that (i) the QD is just a strained region of a high-quality QW with no contaminated interfaces, (ii) the QD PL has an extremely high efficiency, (iii) the decay time of the QD PL is longer than that of the QW we assume that $\tau_{\mathrm{rec}} = 860$ ps is the optical lifetime. This assignment can be further verified by measuring the temperature dependence of the PL decay rate, which is found to be independent of temperature up to ≈ 80 K. The PL decays of the higher-energetic QD transitions ($i = 2,3,4$) are faster than that of the 1-1 transition. As shown by the following model calculations this is due to carrier relaxation from higher-energetic QD levels to lower-energetic QD levels mediated by 0D-2D Coulomb scattering (as long as carriers are present in the QW) and by acoustic phonon scattering. Due to energetic reasons scattering with longitudinal optical (LO) phonons cannot be responsible for the carrier relaxation between the lowest QD levels.

We now present a more quantitative analysis using a simplified model in order to understand the main microscopic processes leading to the observed PL transients. The PL intensities P_i of the transitions i-i are proportional to the mean number $N_{eh,i}$ of respective electron-hole pairs. Since holes are probably present in the dots prior to the optical excitation pulse, we assume in our model that P_i is only proportional to the respective mean number N_i of conduction-band electrons. This simplification is partly justified even in the absence of these extra holes, since the thermalization of the holes should be faster than that of the electrons due to the lower quantization energies of the QD levels in the valence band.[5] To calculate the temporal evolution of $N_i(t)$, we need to know the recombination and relaxation times. As already argued above the optical recombination lifetime $\tau_{\mathrm{rec}} = 860$ ps of the 1-1 transition can be determined without solving a complete rate equation system. Next, the calculations of Ref. 17 predict almost identical optical recombination rates for the four lowest i-i transitions. Regarding relaxation of electrons from the $i = 2$ level to the lowest QD level $i = 1$ we assume that the respective scattering rate τ_{rel}^{-1} is mainly given by the sum of the scattering rates for acoustic phonon scattering (τ_{ph}^{-1}) and for 0D-2D Coulomb scattering. Here the 0D-2D Coulomb scattering rate is considered to be proportional to the 2D carrier density in the uncovered $In_xGa_{1-x}As$ QW. Since the QW PL intensity $P_{\mathrm{QW}}(t)$ is a measure of the carrier density in the QW, we can write for this scattering rate $\tau_{\mathrm{0D\text{-}2D}}^{-1}(t) = R \cdot P_{\mathrm{QW}}(t)$ with R a parameter describing the Coulomb interaction strength. For electron relaxation from higher-lying ($i \geq 3$) QD levels we make the following simplification. We only consider electron relaxation from level i to the next lower-lying QD level $i-1$. However, we do take into account that the number of final scattering states in the lower-lying QD level $i-1$ and thus the scattering rate increases with increasing level index i:

$$\tau_{\mathrm{rel},i}^{-1} = \frac{D_{i-1}}{D_1}\,\tau_{\mathrm{rel}}^{-1} = (i-1)[\,\tau_{\mathrm{ph}}^{-1} + R \cdot P_{\mathrm{QW}}(t)]. \qquad (1)$$

The complete rate equation system then writes

$$\frac{dN_i(t)}{dt} = -\frac{N_i(t)}{\tau_{\mathrm{rec}}} + \frac{N_{i+1}(t)}{\tau_{\mathrm{rel},i+1}}\,\frac{D_i - N_i(t)}{D_i} - \frac{N_i(t)}{\tau_{\mathrm{rel},i}}\,\frac{D_{i-1} - N_{i-1}(t)}{D_{i-1}}. \qquad (2)$$

The factor $[D_i - N_i(t)]/D_i = f(t)$ is a state filling factor taking care of the fact that electrons can only relax to a lower-lying *nonoccupied* dot state (Pauli blocking). We start the calculation of $N_i(t)$ at $t_0 = 0.25$ ns, where the electron densities $N_i(t_0)$ of the five lowest QD levels are taken from the absolute PL intensities (see discussion above). Then only two unknown parameters are left, namely, the acoustic phonon scattering time τ_{ph} from level 2 to 1 and the factor R or $\tau_{\mathrm{0D\text{-}2D}}$ for a given time. Figure 2(b) shows the calculated $N_i(t)$ transients for $i = 1,2,3,4$ when choosing $\tau_{\mathrm{ph}} = 570$ ps and $\tau_{\mathrm{0D\text{-}2D}} = 1$ ps at $t_0 = 0.25$ ns. Obviously, these calculated $N_i(t)$ transients show all the important features of the measured P_i transients supporting the above made model assumptions.

We note that the chosen value of 570 ps for the acoustic phonon scattering time τ_{ph} from $i = 2$ to $i = 1$ is a sensitive parameter in the fit procedure, since this one parameter determines the decay dynamics of $N_i(t)$ for $i = 2$, 3, and 4. Due to the increase of τ_{rel}^{-1} with increasing i [see Eq. (1)] the decay of the $N_i(t)$ transients becomes faster for higher i values. This is also observed for the respective PL transients in Fig. 2(a). The value of 570 ps should be compared to the relatively large quantization energy $\hbar\omega_e = 11$ meV. Very recently, Gammon *et al.*[20] found a much shorter acoustic phonon $2 \rightarrow 1$ relaxation time of 19 ps for a QD sample with a conduction-band quantization energy of less than 2 meV. All this is consistent with theoretical considerations of acoustic phonon scattering in QD's that predict an increase of the scattering time τ_{ph} with increasing quantization energy $\hbar\omega_e$.[5]

The fitted value for the inter-dot-level relaxation time induced by Coulomb scattering is of the order of 1 ps right after the optical excitation. This is consistent with the assumption made above that Coulomb scattering might be responsible for the efficient initial carrier relaxation and thus for the fast onset of the QD PL transients. To show its influence on the PL transients at later times we repeat the model calculations with the same parameters as for Fig. 2(b), but now omitting the term for Coulomb scattering by setting $R = 0$. The results are shown in Fig. 2(c). The initial decrease of the $N_i(t)$ transients, which in a linear plot looks quite pronounced, is not observed in the PL transients of Fig. 2(a) or in the $N_i(t)$ transients of Fig. 2(b), where Coulomb scattering is taken into acccount. This shows that the term for Coulomb scattering is needed to explain the correct shapes of the QD PL transients as long as carriers are in the QW.

To further illustrate the importance of the state filling factor $f(t)$ we now disregard state filling effects by setting $f \equiv 1$ but otherwise using the same parameters as for the calculated $N_i(t)$ transients shown in Fig. 2(c). Figure 2(d) shows the results of such calculations. Obviously, the pla-

teaulike behavior is completely lost and the $N_i(t)$ transients are far from correctly describing the observed QD PL transients of Fig. 2(a).

Finally, we note that our model also describes the QD PL transients for lower excitation intensities correctly (not shown here). As mentioned above a lowering of the excitation intensity mainly leads to a shortening of the initial plateaulike part of the QD PL transients leaving all other features unchanged. This is exactly what we obtain for the $N_i(t)$ transients when we start with smaller numbers $N_i(t_0)$ for the initial mean population of the respective QD levels.

In summary, the temporal dynamics of the photoluminescence from a high-quality QD sample has been investigated by comparing experimental results with those obtained by simple model calculations. The characteristic roles of state filling, and Coulomb and acoustic phonon scattering for the carrier relaxation scenario in a zero-dimensional semiconductor system have been clarified qualitatively and to a certain extent also quantitatively. In the future, more elaborate calculations are needed to understand all the details of the relaxation scenario.

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