Decay dynamics of neutral and charged excitonic complexes in single InAs/GaAs quantum dots

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Systematic time-resolved measurements on neutral and charged excitonic complexes (X, XX, X+, and XX+) of 26 different single InAs/GaAs quantum dots are reported. The ratios of the decay times are discussed in terms of the number of transition channels determined by the excitonic fine structure and a specific transition time for each channel. The measured ratio for the neutral complexes is 1.7 deviating from the theoretically predicted value of 2. A ratio of 1.5 for the positively charged exciton and bieexciton decay time is predicted and exactly matched by the measured ratio indicating identical specific transition times for the transition channels involved.

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than XX and XX+, suggesting a feeding process from an external charge carrier reservoir rather than an intrinsic feeding. Such slow components were previously controversially attributed to reemission and lateral transfer of charge carriers [13, 14] or to conversion of dark excitons to bright excitons via spin-flip processes [15]. For the X+, XX, and XX+ decays, the spin flip can be ruled out as these complexes possess no such dark states. Instead, the source of the feeding process is suggested to be outside of the QDs, e.g., neighboring shallow defect states or reemission from other QDs. In the following, we will focus on the dominant fast component.

Figure 2 shows the dominant decay times of X, X+, XX, and XX+ as a function of the X+ recombination energy. A clear sequence of decay times for the excitonic complexes is obvious. X exhibits the longest decay time followed by X+, XX, and finally XX+, analogous to Fig. 1 and in agreement with previous reports on InAs/GaAs QDs [3, 4].

No clear trend for a dependence of the decay times on the X+ energy is identified. Apparently, the structural parameters, which lead to differences in the transition energies, do not control the decay times in the same way. Interestingly, the scatter of the X decay times is much larger than that of the other excitonic complexes. The mean decay times are $\tau(X) = 1.22 \pm 0.25$ ns, $\tau(X+) = 0.97 \pm 0.15$ ns, $\tau(XX) = 0.76 \pm 0.12$ ns, and $\tau(XX+) = 0.66 \pm 0.14$ ns. The standard deviation of $\tau(X)$ is almost twice as large as for the other complexes. The X wave function seems to be more sensitive to the structural properties of the QD than the wave functions of complexes containing more holes.

The decay times of the neutral and charged complexes will now be compared with respect to the number of allowed transitions of each excitonic complex.

The radiative decay time $\tau(EC)$ of an excitonic complex EC is composed of the specific transition time $\tau_{i,f}$ of all recombination channels involved [10]:

$$\frac{1}{\tau(EC)} = \sum_{i,f} n_i \frac{1}{\tau_{i,f}},$$

with the mean population $n_i$ ($\sum n_i = 1$) of the initial state. It is assumed that the STTs for different transition channels ($|i\rangle \rightarrow |f\rangle$ and $|i'\rangle \rightarrow |f'\rangle$) of one complex are identical ($\tau_{i,f} = \tau_{i',f'} = \tau_{EC}$) since the oscillator strength is independent of the actual spin configuration as long as the spin selection rules are obeyed. Under this precondition, the expected excitonic decay times $\tau(X)$, $\tau(X+)$, $\tau(XX)$, and $\tau(XX+)$ are determined by the number of allowed recombination channels and their STTs, respectively.

Figure 3 shows the energy schemes of the four excitonic complexes to illustrate the different transition channels. The initial and final states are labeled with their respective total spin configurations.
The initial configuration of the optically active X consists of the two bright states with the spin configuration $| -1 \rangle \pm | 1 \rangle$. The dark states $| -2 \rangle \pm | 2 \rangle$ are optically forbidden and do not contribute to the recombination process. The exciton fine structure splitting of the QDs probed here was determined to be less than 20 µeV. Hence, the mean initial population $n_{\pm 1}$ of the two bright states is $\frac{1}{2}$. Each state possesses one recombination channel to the ground state $| 0 \rangle$. The decay time then amounts to $\tau(X) = \tau_X$.

In contrast to X, XX consists of a single initial state $| 0 \rangle$ with $n_0 = 1$ and possesses two recombination channels to the two final states: the bright states of the X ($| -1 \rangle \pm | 1 \rangle$). Hence, the decay time amounts to $\tau(X) = \frac{1}{2} \tau_{XX}$.

To compare the decay times of X and XX, identical STTs for both complexes are assumed. Narvaez \textit{et al.} calculated a size dependent ratio between 1.5 and 2 [8], which is similar to the measured ratio. Just like for the neutral complexes, equal STTs ($\tau_{X+} = \tau_{XX+}$) for the charged complexes are assumed. Since the measured ratios of the X and XX decay times (black squares) and of the X+ and XX+ decay times (red circles) are plotted as a function of X+ recombination energy, the decay times are governed mainly by $\tau(X)$, thus, supporting our measurements.

The measured mean value of $\tau(XX)$ is 1.7 with a standard deviation of 0.4. For $\tau(X+)$, the mean value results to 1.5 with a standard deviation of 0.2.

The mean $\tau(XX)$ value is clearly below the predicted value of 2 with a large scatter of the individual values. Qualitatively, the argument based on the number of involved transition channels holds. The exact value, however, crucially depends on the specific QD since the STTs differ a lot from case to case and are not always comparable as in [10]. Wimmer \textit{et al.} calculated a size dependent ratio between 1.5 and 2 [8]. The change in the ratio is governed mainly by $\tau(X)$, thus, supporting our measurements.

The measured mean value of $\tau(XX)$ matches exactly the expected value of 1.5. The scatter is considerably smaller than for $\tau(X)$ with a standard deviation of 0.2 indicating a stronger similarity of the STTs of X+ and XX+ from dot to dot than for the neutral complexes. Here, the number of allowed transitions is the key parameter determining the ratio of decay times of both complexes.

The comparison of the measured $\tau(X)$ and $\tau(X+)$ shows that the STTs $\tau_X$ and $\tau_{X+}$ differ strongly. Adding a single charge carrier to an excitonic complex influences the wave functions and, therefore, their overlap is much stronger than adding a neutral pair of charge carriers.

In summary, we presented a systematic study of the decay dynamics of excitonic complexes in 26 different single InAs/GaAs QDs. The influence of the specific transition times and electronic fine structure on the dynamics was analyzed. Surprisingly, the scatter of the neutral exciton’s decay time from dot to dot is larger than the scatter of the other complexes’ decay times indicating a
greater sensitivity of the exciton wave function to details of the structural properties of the QD. No dependence on the transition energy was observed in the 1.26 – 1.35 eV range. The mean value of $\tau(X)/\tau(XX)$ was 1.7 ± 0.4, thus, below the value of 2 as given by the different number of decay channels alone. This implies shorter STTs for X than for XX. For $\tau(X+)/\tau(XX+)$ a value of 1.5 was predicted. The mean measured ratio is 1.5 ± 0.2, thus, matching the expected theoretical value. This indicates similar STTs for the transition channels of X+ and XX+. The additional positive charge carrier seems to stabilize the wave function overlap when an additional exciton is added to the complex.

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