Excitonic lifetimes in single GaAs quantum dots fabricated by local droplet etching

Ch Heyn\textsuperscript{1,3}, Ch Strelow\textsuperscript{2} and W Hansen\textsuperscript{1}

\textsuperscript{1} Institute of Applied Physics, University of Hamburg, D-20355 Hamburg, Germany
\textsuperscript{2} Institute of Physical Chemistry, University of Hamburg, D-20146 Hamburg, Germany
E-mail: heyn@physnet.uni-hamburg.de

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Abstract. The time-dependent optical emission of GaAs quantum dots (QDs) is studied using single-dot photoluminescence (PL) spectroscopy with quasi-resonant excitation into the QD d-shell. The QDs are fabricated with a very recently developed method, i.e. by local droplet etching of self-assembled nanoholes in an epitaxial AlAs/AlGaAs heterostructure surface and subsequent filling with GaAs. The PL data are interpreted in terms of a three-level rate model, which yields lifetimes of 390 and 426 ps for the excitons and biexcitons, respectively. The strong dependences of both the PL peak intensities and decay times on the excitation power are quantitatively reproduced by the model. The comparison with various other types of self-assembled QDs shows the trend of a decreasing exciton lifetime with increasing emission energy. This behavior and the short lifetime of the GaAs QDs are discussed on the basis of common models of the exciton radiative lifetime.

\textsuperscript{3} Author to whom any correspondence should be addressed.
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1. Introduction

Semiconductor quantum dots (QDs) featuring simultaneous zero-dimensional confinement for electrons and holes are suggested for fundamental research [1] on charge carrier quantization and interaction as well as for advanced applications in the fields of quantum computing [2] and quantum cryptography [3, 4]. A very interesting topic in this context is excitons residing inside single QDs and their dynamic behavior [1]. Time-resolved measurements of the exciton (X) and biexciton (XX) optical emission have been carried out for various well-known QD systems, such as e.g. CdSe QDs [5–7], InGaAs QDs [8–12], InGaN QDs [13] and GaAs monolayer fluctuation QDs [14]. In particular, QD systems with high confinement potential and small radiative lifetime are required, for instance for single-photon sources with high repetition rate and fast optical spin readout.

The present work discusses the excitonic time evolution of a novel type of GaAs QDs fabricated by a very recently developed technique. Nanoholes in an epitaxial AlAs/AlGaAs heterostructure surface are generated with local droplet etching (LDE) [15–18]. LDE is fully compatible with molecular beam epitaxy (MBE) and forms patterned semiconductor surfaces without the need of any lithographic steps. Strain-free GaAs QDs are created by partial refilling of LDE nanoholes in AlAs or AlGaAs surfaces with GaAs [19–21]. A cross-section of an LDE QD is shown in figure 1.

The excitonic time evolution is measured using time-resolved single-dot photoluminescence (PL) spectroscopy under pulsed excitation [5–14]. After the laser pulse, the PL emission from the QD levels appears to follow a multi-exponential decay. The decay times obtained from single-exponential fits of the data do not reflect the true excitonic lifetimes. Single-exponential decay times are expected only for single-particle single-level systems, but not for complex QDs with several coupled levels. Therefore, the excitonic dynamics is often analyzed by using rate-equation-based models that allow the calculation of the time-dependent occupation probabilities of the various levels inside a QD [5, 8]. An often used simple model is the two-level approach of Bacher et al [5]. More levels are considered in the multi-level approach proposed by Dekel et al [8] In this paper, the PL data are analyzed using a three-level rate model of the QD excitonic states similar to the approach of Dekel et al but under consideration of quasi-resonant excitation into the QD d-shell. The obtained exciton and biexciton lifetimes are compared to those of other QD systems.
Figure 1. Low-temperature PL spectrum of a GaAs QD ensemble showing the s–s peak at 1.576 eV, the p–p peak at 1.632 eV and the d–d peak at 1.690 eV. The excitation energy is $E_E = 2.33$ eV. The left inset shows a typical single-dot spectrum of the sample with neutral exciton (X) and biexciton (XX) peaks and the right inset depicts a schematic QD cross-section.

2. Sample fabrication and single-dot measurements

The GaAs QD fabrication is performed in a conventional solid-source MBE system. The process starts with the MBE growth of an AlGaAs buffer with Al content of 0.36 on a (001) GaAs substrate. This is followed by a 5 nm thick AlAs layer, which is important for the size uniformity [19, 21] of the later nanoholes. Now the As flux is switched off and 3.2 monolayers of Al are deposited at a temperature of 650 °C. Thereby Al droplets are formed on the surface in the Volmer–Weber growth mode. During a subsequent 180 s annealing step, the droplets are transformed into nanoholes [17, 18]. For QD generation, the nanoholes are refilled with GaAs. Hereby, the QD size is precisely adjusted by the filling level [19]. For the present QDs, deposition of a 0.565 nm thick GaAs layer produces QDs with a nominal height of 7.6 nm. Finally, the QDs are capped with a 120 nm thick AlGaAs barrier. More details of the QD fabrication technique by utilizing the self-assembled LDE can be found in [19–21], and a cross-sectional transmission electron microscopy study of a similar dot sample can be found in [22]. The dot density can be adjusted to be very low and is about $4 \times 10^5$ cm$^{-2}$ in the sample discussed here. Thus, for single-dot PL it is sufficient to focus the excitation laser beam to a spot of diameter less than 1 µm.

Figure 1 shows an overview of the optical emission from the QD sample with ensemble PL measurements of the shell structure and a typical single-dot spectrum revealing the excitonic features of the s-shell. According to the conventional nomenclature, the electron and hole ground states in the shell picture are denoted by s-, the first excited by p- and the second excited by d-states. Due to optical selection rules, radiative recombinations take place mainly between electrons and holes in states of equal quantum index and are denoted here by s–s, p–p and d–d.
transitions. Due to spin degeneracy, an s-state can be occupied by a maximum of two electrons or holes. This yields four different types of s–s transitions related to recombinations of neutral excitons, neutral biexcitons, positively charged excitons, i.e. excitons in the presence of a hole, and negatively charged excitons, i.e. excitons in the presence of an electron. A typical single-dot spectrum of the s–s transitions shows the neutral exciton peak (X) at an energy that is about 1.3 meV higher compared to the biexcition peak (XX) (figure 1). Both peaks have been identified by their excitation power dependence as has been described previously [21]. The additional peaks are probably related to charged excitons.

A Ti-sapphire laser is used for the measurements, allowing quasi-resonant excitation into the QD d-shell. An optical oscillator pumped by the Ti-sapphire laser is used for excitation into the AlGaAs barrier material. For time-dependent measurements, a laser pulse of 250 fs is applied at a repetition frequency of 75.4 MHz. The PL emission from the QD is measured using a Hamamatsu C5680 streak camera with an MC5675 Synchroscan plug-in and a time resolution of approximately less than 30 ps together with a 30 cm spectrometer.

In the first set of time-resolved experiments, we have used a laser with an excitation energy of $E_E = 2.54$ eV, which generates excitons inside the AlGaAs barrier material. Here, the exciton and biexciton PL peaks show complex decay behavior, which cannot be fitted by a simple single-exponential decay. Furthermore, the typical decay times are several nanoseconds. Qualitatively different behavior is observed with quasi-resonant excitation into the QD d–d transition using a laser energy of $E_E = 1.69$ eV. As is demonstrated in figure 2(c), now the excitonic PL lines follow a single-exponential decay and the decay times are about 500 ps. We explain this difference with a reservoir effect induced by the large number of states inside the AlGaAs barrier material feeding excitons into the QD during non-resonant excitation.
This reservoir effect interferes with the time dependence of the QD optical emission and thus dominates measurements of the QD excitonic lifetimes. Therefore, all the following experiments are performed under resonant excitation into the QD d–d transition.

Figure 2(a) shows an example of the raw data from the streak camera at an excitation intensity of \(I_E = 7 \mu W\). We attribute the intensive horizontal line at \(t \approx 0\) to hot excitons [23] being emitted from the GaAs substrate during and very shortly after the laser irradiation. After decay of the hot exciton emission, one intensive vertical line at a wavelength of 780.0 nm remains accompanied by several weaker lines at higher wavelengths. These lines are also visible in the corresponding PL spectra in figure 2(b) recorded at \(t = 300\) ps. The energetic separation between the strongest and the second strongest peaks in figure 2(b) is about 1.3 meV, which is identical to the exciton–biexciton splitting observed under cw illumination (figure 1). We identify the peaks in the PL spectra by comparison with single-dot spectra of the same sample obtained under cw illumination and varying excitation intensity [21]. Accordingly, we label the strong peak with the highest energy in figure 2(b) as exciton (X) and the second peak as biexciton (XX). Our identification is supported by the results of k.p calculations yielding a red-shifted biexciton with respect to the exciton [24], as well as by polarization-dependent measurements on a similar dot sample [21]. Furthermore, figure 2(b) shows a PL spectrum from the same QD but at a significantly reduced excitation intensity \(I_E = 750\) nW. There, the exciton peak is visible and the biexciton is below the noise level.

The time evolution of the exciton peak maximum at 1.588 eV is plotted in figure 2(c). For \(I_E = 7 \mu W\), the laser pulse generates in the substrate a strong hot-exciton-related spike at \(t = 0–50\) ps, as is described above. After excitation, the exciton peak intensity first strongly increases for about 300 ps and afterwards decays according to a single-exponential function. As demonstrated with the \(I_E = 750\) nW data in figure 2(c), this behavior is also visible at low excitation intensity.

3. Rate model of the excitonic time evolution

The applied three-level rate-model of the QD excitonic time evolution considers the QD d-shell and the s-shell with neutral exciton and biexciton levels. A scheme is shown in figure 3(a). The nomenclature in terms of the QD shell structure is based on the assumption that excitons are formed by electron and hole states of the same quantum index.

Bright excitons are generated with an excitation rate \(R_E\) by a 250 fs laser pulse with energy that is resonant with the QD d-shell. Dark states are not considered in the model, since the bright exciton radiative lifetimes are expected to be much shorter than spin relaxation times into the dark state [5]. \(R_E\) is given in units of the number of generated excitons per ns and QD. The laser irradiation increases the exciton occupation probability \(N_d\) in the d-shell.

After a lifetime \(\tau_d\), excitons from the d-shell relax into the QD s-shell. The s-shell is either empty or filled with one exciton or with two excitons (denoted as one biexciton). The corresponding time-dependent occupation probabilities are \(N_X\) and \(N_{XX}\), respectively. The probability of an empty s-shell is \(1 - N_X - N_{XX}\). Thus, relaxation of d-shell excitons into an empty s-shell takes place with rate \((1 - N_X - N_{XX})N_d/\tau_d\) and increases the occupation probability \(N_X\). Relaxation of d-shell excitons into an s-shell already filled with one exciton takes place with rate \(N_X N_d/\tau_d\). This process decreases \(N_X\) and increases the biexciton occupation probability \(N_{XX}\). This yields a total rate of exciton relaxation from the d-shell of \((1 - N_{XX})N_d/\tau_d\). The QD s-shell is completely filled when occupied with one biexciton. Other
levels such as the p-shell are not considered in the model in order to keep the number of free parameters low.

After a lifetime \( \tau_X \), an exciton recombines under emission of one photon. In the case of a biexciton, we assume that after a lifetime \( \tau_{XX} \) only one of the two excitons recombines under photon emission and that the second exciton increases \( N_X \).

Based on this approach, the time-dependent occupation probabilities of the d-shell and of the exciton and biexciton levels inside the s-shell are described by the following set of coupled rate equations:

\[
\frac{dN_d}{dt} = R_E - (1 - N_{XX}) \frac{N_d}{\tau_d},
\]

\[
\frac{dN_X}{dt} = (1 - 2N_X - N_{XX}) \frac{N_d}{\tau_d} + \frac{N_{XX}}{\tau_{XX}} - \frac{N_X}{\tau_X},
\]

\[
\frac{dN_{XX}}{dt} = N_X \frac{N_d}{\tau_d} - \frac{N_{XX}}{\tau_{XX}}.
\]

As a proof of the model consistency, the balance between incoming and emitted photons is conserved by equations (1)–(3): \( dN_d/dt + dN_X/dt + 2dN_{XX}/dt = R_E - N_X/\tau_X - N_{XX}/\tau_{XX} \).

The measured PL intensities are proportional to the radiative recombination rates \( R_e = N_X/\tau_X \) for excitons and \( R_R = N_{XX}/\tau_{XX} \) for biexcitons, respectively. In a first step, we have determined the exciton lifetimes \( \tau_d, \tau_X \) and \( \tau_{XX} \). Equation (3) shows that after emptying the d-shell at \( t \geq 500 \) ps (figure 3(c)) the biexciton occupation probability follows a simple single-particle behavior. Therefore, here the PL decay time is identical to the radiative lifetime.
This allows for a simple determination of $\tau_{XX} = 426$ ps. The remaining lifetimes $\tau_d = 108$ ps and $\tau_X = 390$ ps are determined by adjusting them with respect to the best reproduction of the experimental time-dependent exciton and biexciton PL emission at $I_E = 3.7 \, \mu W$. Figure 3(b) shows that there is good quantitative agreement. The excitation rate of $R_E = 740$ excitons per ns, yielding a ratio $R_E/I_E = 200$ excitons per ns and $\mu W$, is also determined by fitting. This ratio and the above lifetimes are kept constant for all calculations. Furthermore, the ratio between the calculated radiative recombination rates and the measured PL counts is also assumed to remain constant.

4. Influence of the excitation rate

As a test of the model assumptions, we have calculated the time evolution of the exciton and biexciton radiative recombination rates under cw illumination. Two examples for $R_E = 0.2 \, ns^{-1}$ and $R_E = 2 \, ns^{-1}$ are shown in figures 4(a) and (b). In all cases, we find an initially very strong increase of $R_R$, which is followed by saturation at a constant level. Furthermore, the dependence of the saturated exciton and biexciton recombination rates on the excitation rate is plotted in figure 4(c). The model results show that at small $R_E$ the exciton recombination dominates with an approximately linear increase. This is followed by a decrease at higher $R_E$. The biexciton recombination rate is very small at low $R_E$, but increases parabolically and surpasses the excitons. These results are in agreement with the widely accepted behavior [25], which confirms the validity of the present rate model.

In the next step, we have calculated the exciton and biexciton recombination rates for pulsed excitation during $t = 0–250$ fs. In figures 5(a)–(c), a comparison between the model results and experimental PL intensities is plotted for two QDs and varied excitation conditions. Since both QDs have nearly identical exciton PL peak energies of 1.590 eV (dot 1) and 1.588 eV
Figure 5. (a)–(c) Comparison of the time evolution of the exciton (X) and biexciton (XX) peak intensities (thin lines) with model results (thick lines) under pulsed excitation for 250 fs at varied excitation conditions as indicated. The model is parameterized for best agreement with the $I_E = 3.7 \, \mu W$ data of (b). Other calculations are performed using the same model parameters. The ratio $R_E/I_E = 200$ excitons per ns and $\mu W$ as well as the ratio between calculated recombination rates and measured PL counts are also kept constant. (d) Comparison of the calculated maximum $R_R$ with the measured maximum PL intensity as a function of the excitation intensity for two different QDs. (e) Comparison of the decay times $\tau_S$ obtained by fitting the experimental data (symbols) and model results (lines) using a single-exponential function.

(dot 2), we assume that the excitonic lifetimes of both QDs are also very similar. For $R_E$ up to $740 \, \text{ns}^{-1}$ we find very good quantitative agreement, whereas for higher $R_E$ the calculated exciton recombination rates are smaller in comparison to the experimental data. This is also visible in figure 5(d), which compares the maximum measured PL intensities and calculated recombination rates. The model predicts a saturation of the maximum exciton recombination rate at higher $R_E$, which is not clearly observable in the experiments. This discrepancy for high $R_E$ might be caused by the onset of p-shell-related recombinations (not considered in the model). In contrast to that, the biexciton PL intensities are quantitatively reproduced by the model even at high $R_E$.

Furthermore, the exciton and biexciton decay times are analyzed by fitting experimental data and calculation results with a single-exponential function. The results are shown in figure 5(e). We find a strong increase of the exciton decay times for both the experiments and the model results. The smaller slope of the experimental data might be related to a p-shell-induced reservoir effect, which is not considered in the model. For the biexcitons, there is only a weak influence of the excitation conditions. This is in agreement with the predictions of equation (3) for small $N_d$ (figure 3(c)).
Figure 6. (a) Exciton lifetimes $\tau_X$ measured (Exp) for different types of self-assembled QDs as indicated together with values calculated with two different models (model 1 and model 2) as described in the text. Lines are a guide to the eyes. As indicated by the dotted line, in the strong confinement regime, i.e. $E_X < 1.4 \text{ eV}$, the values of model 2 are larger than the time range depicted. (b) Ratio of the bulk exciton Bohr radius $\lambda_B$ and the QD radius $r_{QD}$ for different QD types of (a). The values used for the calculations are listed in table 1.

5. Discussion and conclusions

The exciton radiative lifetime $\tau_X = 390 \text{ ps}$ determined here for the GaAs QDs is compared in figure 6(a) with the values obtained for other QD systems. Shorter lifetimes are found for CdTe and CdSe QDs, and longer ones for InAs, InGaAs, InP and InGaN QDs. The short radiative lifetime suggests the present GaAs QDs, for instance, for single-photon sources with a high repetition rate and a fast optical spin read-out. Regarding the trend of the lifetime data, one finds that the values of $\tau_X$ decrease with increasing exciton emission energy $E_X$. Only the InGaN QDs deviate from this trend.

In order to identify the parameters controlling $\tau_X$, we apply a simple model of the exciton lifetime in the single-particle limit [12]:

$$\tau_X = \frac{3 h^2 c^3 \epsilon_0 m_0}{n\pi e^2 E_X E_P \langle \psi_e | \psi_h \rangle^2}$$

with the refractive index $n$, the Kane energy $E_P$ and the overlap integral $\langle \psi_e | \psi_h \rangle^2$ between the electron and hole envelope wave functions in the QD. In this approach (denoted by model 1) we assume that the overlap integral may be calculated from uncorrelated single-particle QD states in the strong confinement limit. If the electron and hole are localized in roughly the
Table 1. References for the experimental lifetime data of figure 6(a) together with the values of the exciton emission energy $E_X$, the QD height $h_{QD}$, the QD radius $r_{QD}$, the Kane energy $E_P$, the refractive index $n$ and the bulk exciton Bohr radius $\lambda_B$.

| QD type | References | $E_X$ (eV) | $h_{QD}$ (nm) | $r_{QD}$ (nm) | $E_P$ (eV) | $n$ | $\lambda_B$ (nm) |
|---------|------------|------------|--------------|--------------|-------------|-----|-----------------|
| InAs    | [26]       | 0.94       | 2            | 17.5         | 21.1 [26]   | 3.9 | 31.6            |
| InAs    | [10]       | 1.05       | 13           | 24.5         | 21.1 [26]   | 3.9 | 31.6            |
| InGaAs  | [12]       | 1.3        | 2.5          | 14           | 21.1 [26]   | 3.9 | 17.5            |
| GaAs    | The present work | 1.59 | 7.6          | 20           | 22.7 [29]   | 3.6 | 11.7            |
| InP     | [27]       | 1.75       | 7            | 35           | 20.7 [30]   | 3.4 | 9.3             |
| CdTe    | [28]       | 2          | 5            | 15           | 17.9 [28]   | 3   | 6.8             |
| CdSe    | [5]        | 2.69       | 1            | 5            | 16.5 [31]   | 2.7 | 1.3             |
| InGaN   | [13]       | 2.83       | 4.6          | 5.8          | 15.7 [32]   | 2.4 | 4.4             |

In the same way, we have maximum overlap, i.e. $\langle \psi_e | \psi_h \rangle^2 \simeq 1$. Model 1 establishes an upper bound on the radiative lifetime. Exciton lifetimes calculated with model 1 for different QD types are plotted in figure 6(a) together with the experimental values. The calculated lifetimes reproduce well the experimental trend of decreasing $\tau_X$ with increasing $E_X$. On the other hand, model 1 overestimates the experimental lifetimes, in particular for higher $E_X$. We assume that the discrepancy for high $E_X$ is related to correlation effects that are not included in equation (4). Correlation is expected to become important if the dot size becomes larger than the exciton Bohr radius. This is supported by figure 6(b), where the ratio of the bulk exciton Bohr radius $\lambda_B = 4\pi \epsilon_s \epsilon_0 \hbar^2/(m^*_e e^2)$ to the QD radius $r_{QD}$ is plotted. Here $\epsilon_s$ is the semiconductor dielectric constant, $m^*_e = (1/m^*_e + 1/m^*_hh)^{-1}$ the exciton effective mass, $m^*_e$ the electron and $m^*_hh$ the heavy-hole effective mass. Strong and weak confinement are expected for $\lambda_B/r_{QD} \gg 1$ and $\lambda_B/r_{QD} \ll 1$, respectively.

In order to address excitonic effects in the weak confinement regime, we have multiplied the lifetimes of equation (4) by a factor $(\lambda_B/r_{QD})^2$. Corresponding lifetimes are marked by open triangles in figure 6(a) and reproduce the trend of the experimental data for QD systems in the weak confinement regime. Naturally, in the strong confinement regime, the thus obtained lifetimes are much too high (out of the range of the $\tau_X$ axis in figure 6(a)). Importantly, the present GaAs QDs lie in the transition regime between strong and weak confinement. This enables the interesting possibility of tuning these QDs from weak to strong confinement via the controlled variation of the QD size [19].

Furthermore, our data reveal a biexciton radiative lifetime that is slightly higher compared to the lifetime of the exciton. In the simplest argumentation [10], a biexciton would act like two independent excitons with doubled recombination rate and, thus, $\tau_X/\tau_{XX} = 2$. However, considering the Coulomb interaction between electrons and holes inside the QD, Bacher et al [5] estimated a ratio $\tau_X/\tau_{XX} \simeq 1$, which is close to the present experimental results.

Figure 5(e) indicates an increase of the exciton decay time $\tau_S$ with increasing intensity $I_E$. This behavior does not reflect an increase of the exciton lifetime $\tau_X$, but instead originated from a reservoir effect of the biexcitons induced by the relative increase of the biexciton occupation probability with respect to the excitons (figure 5(d)). On the other hand, the biexciton decay time increases only slightly with $I_E$ and its values are close to the lifetime.

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A great advantage of the present GaAs QDs is the possibility of varying their size in a very controlled fashion by the nanohole filling level \[19\]. Furthermore, the present QDs are strain-free and their composition is not influenced by unintentional intermixing processes during growth. This perfect control on the QD structural properties allows, for instance, detailed studies of the correlation between the excitonic lifetimes and the size of the confining potential.

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