

# Excitation mechanisms of individual CdTe/ZnTe quantum dots studied by photon correlation spectroscopy

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Systematic measurements of auto- and cross-correlations of photons emitted from individual CdTe/ZnTe quantum dots under pulsed excitation were used to elucidate nonresonant excitation mechanisms in this self-assembled system. Memory effects extending over a few excitation pulses have been detected in agreement with previous reports and quantitatively described by a rate equation model, fitting a complete set of correlation and PL intensity results. The important role of single carrier trapping in the quantum dot was established. An explanation was suggested for the unusually wide antibunching dip observed previously in  $X$ - $X$  autocorrelation experiments on quantum dots under cw excitation.

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## I. INTRODUCTION

The emerging field of quantum information has given rise to an interest for sources of photons-on-demand. Semiconductor light sources offer important advantages, such as low power consumption and possibilities of integration with existing electronics. Precise knowledge of quantum dot (QD) excitation mechanisms is of primary importance for the creation of semiconductor sources of photons-on-demand. In CdTe/ZnTe QDs, some aspects of nonresonant excitation are not yet fully understood, e.g., long components of photoluminescence (PL) decay<sup>1</sup> and the unusually wide antibunching dip in autocorrelation of photons from exciton recombination<sup>2</sup> (excitonic photons). For the prototypical InAs/GaAs system, it has been established by Santori *et al.*<sup>3</sup> that among the many memory processes in QD optical excitation, medium time scale (sub  $\mu$ s) blinking leads to the enhancement or suppression of excitonic photon autocorrelation for resonant or nonresonant QD excitation, respectively. The QD charge state variation was suggested as the source of these effects. We present here a systematic study of nonresonant excitation mechanisms by photon correlation spectroscopy of CdTe/ZnTe single quantum dots.

## II. SAMPLE AND CHARACTERIZATION

The sample was grown by molecular beam epitaxy on a (001) oriented GaAs substrate, as described in Ref. 4. A 4  $\mu$ m CdTe buffer layer was followed by a 100 nm ZnTe barrier. Then two monolayers of CdTe were grown, forming fluctuation-type quantum dots. The QD layer was overgrown by a ZnTe barrier of 50 nm thickness. Transmission electron microscopy measurements performed on such samples revealed quantum dots with a typical lateral size of 3 nm and a density of  $10^{12}$  cm<sup>-2</sup> (Ref. 4).

The basic characterization was performed by standard cw photoluminescence (PL) excited with an Argon ion laser and by time-resolved photoluminescence excited at 3.5 eV by

frequency-doubled 150 fs pulses from a Ti:sapphire laser. In the time resolved characterization a streak camera (resolution of about 10 ps) was used to record spectral and temporal distribution of PL. A typical PL spectrum is presented in Fig. 1(a). Three major components can be distinguished. The set of three lines at the highest energy (2.345–2.385 eV) is related to the exciton emission from the barrier material (ZnTe). The middle energy component (2.30–2.34 eV) is due to the wetting layer, and the one at the lowest energy (2.20–2.30 eV) is related to the emission from quantum dots. This natural assignment is consistent with the results of further experiments.

The typical temporal profiles of the barrier and QD lines are presented in Fig. 1(b). The ZnTe emission consists of a fast-decaying ( $\tau_{D1}=18\pm 3$  ps) free exciton line at 2.375–2.385 eV and long-lived lines of acceptor-bound and donor-bound excitons. The fast decay provides information on the decay of free excitons and was used as an approxi-

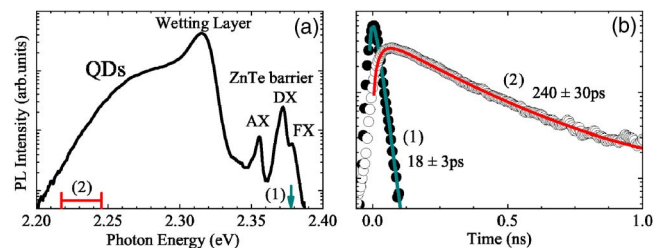


FIG. 1. (Color online) (a) Time-integrated (0–1.6 ns) PL spectrum of the CdTe/ZnTe sample at  $T=7$  K. Structures originating from acceptor-bound (AX), donor-bound (DX), and free (FX) excitons from the ZnTe barrier as well as from excitons in the wetting layer are indicated. Emission from the QD layer forms a wide band centered at 2.25 eV. Markers (1) and (2) indicate energies corresponding to temporal profiles presented in Fig. 1(b). (b) PL transients of free excitons in ZnTe barriers (1) and quantum dots (2) at  $T=7$  K. Points: experimental data and solid lines: fitted curves. Respective decay times indicated.

mate measure of the characteristic time of quantum dot excitation. The decays of the QD PL lasted much longer. The dominant decay time was about  $\tau_{D2}=240\pm 30$  ps, but longer components were also observed. The decay is not straightforward in interpretation. The macro-PL signal is composed of many different lines related to different quantum dots. In particular, lines due to neutral excitons ( $X$ ), biexcitons ( $XX$ ), and trions ( $CX$ ) may decay with different characteristic times. Thus the measurements on single quantum dots were used to determine decay times of particular lines (see Sec. III C). However, the rise times of the QD PL ( $\tau_R=25-30$  ps) were significantly longer than the resolution-limited rise times of the ZnTe PL ( $\tau_R$  about 10 ps) and close to the decay time of the FX line [Fig. 1(b)]. This shows that the QDs are not excited directly by light pulses and points to the transfer of carriers (excitons) from barriers as a source of QD excitation. The characteristic time of this transfer will be used in the rate equation model presented in Sec. IV A. The same values have been determined by Viale *et al.*<sup>5</sup> in a PL dynamics study of a similar CdTe/ZnTe QD system, and attributed to relaxation processes from excited to the ground QD exciton state. As we will see in Sec. IV B, such an interpretation cannot be maintained in our case. We select an alternative interpretation following that proposed in Ref. 6 for III-V QDs. It was shown in Ref. 6 that the excitation is transferred from carriers photo-created in the barriers to the QD excitons not through excited QD states but through a continuum of background states coupled to the QD ground state.

### III. SINGLE QUANTUM DOT SPECTROSCOPY

In order to achieve a better insight in QD excitation and recombination processes, several different experiments on single QDs were performed such as precise determination of excitonic line energy positions, measurement of QD in-plane anisotropy (not shown), and dependence of QD PL intensity on excitation power (Sec. III B). In order to obtain excitonic radiative lifetimes we measured the decays of individual QD emission lines (Sec. III C). Preliminary excitation correlation spectroscopy (ECS) experiments on individual quantum dots were performed (Sec. III D) to estimate the temporal scale of the QD excitation processes. Correlated photon counting with femtosecond pulsed excitation was also performed (Sec. III E). Both autocorrelation and cross-correlation were measured, providing information on QD excitation mechanisms.

#### A. Experiment

For studies of individual quantum dots, a microphotoluminescence ( $\mu$ -PL) setup was used, assuring a spatial resolution better than 1  $\mu\text{m}$ . The sample was fixed directly on the front surface of a mirror type microscope objective<sup>7</sup> (numerical aperture = 0.7) inside a pumped helium cryostat and cooled down to  $T=1.7$  K. An argon ion laser (488 nm or multiline uv) was used for cw excitation. For time-resolved measurements, frequency-doubled pulses of a mode-locked Ti:sapphire laser were used (repetition frequency of 76 MHz, spectral and temporal width 2.6 nm and less than 1 ps, re-

spectively). Averaged excitation power was varied in a range from 10 nW to 4  $\mu\text{W}$  by use of neutral density filters.

In excitation correlation spectroscopy the PL was excited by pairs of pulses obtained by splitting of the pulsed laser beam. Temporal separation between the two pulses of each pair, controlled by an optical delay line, ranged from 0 to 1.5 ns. Time-integrated spectra were recorded with a charge coupled device (CCD) camera. Single photon correlation measurements were performed in a Hanbury-Brown and Twiss setup with spectral filtering by grating monochromators (spectral resolution 200  $\mu\text{eV}$ ). Each monochromator was equipped with a CCD camera and an avalanche photodiode (APD) serving as a single photon detector (temporal resolution 750 ps, quantum efficiency 55% at 560 nm, dark  $<200/\text{s}$ ). For PL decay measurements one of the APDs was replaced by a microchannel plate (MCP) photomultiplier tube (temporal resolution of 40 ps). A correlation card with a multichannel analyzer (4096 time bins, 146 ps width each) was used to generate histograms of correlated photon detection events versus delay, equivalent to an unnormalized second order correlation function.

The same card was used to record PL decay curves by measuring time distance between photon detection and laser pulse reference signal. The channel width was set to 37 ps in that case.

Certain experiments were performed at doubled pulse repetition frequency. This was achieved by dividing the laser beam in a 50/50 nonpolarizing beamsplitter and delaying one of the components by half (6.6 ns) of the initial repetition period, before combining both components in a single beam.

#### B. Identification of single QD PL lines

Because of the high dot density, individual dot lines could be well-resolved only in the long-wavelength tail of the spectrum. In a previous study on the same sample<sup>8</sup> we found that a typical spectrum of an individual QD contains a neutral exciton line, accompanied by biexciton and charged exciton lines, at about 13 and 11 meV below, respectively. The identification was evidenced by a series of experiments, including synchronized line energy jumps confirming that observed transitions originate from the same spot of the sample,<sup>9</sup> in-plane optical anisotropy, and Zeeman effect measurements.

We selected for further measurements a quantum dot emitting a group of PL lines presented in Fig. 2. Besides the lines corresponding to those identified in Ref. 8, a weak line  $CX'$  appears in the spectrum. By measuring in-plane optical anisotropy, we confirmed equal absolute values and opposite signs of the anisotropic exchange splittings for  $X$  and  $XX$  lines. No measurable anisotropy was obtained for  $CX$  and  $CX'$  lines which allows us to ascribe their origin to recombination of two oppositely charged excitons confined in the chosen dot. In further experiments we focused our attention on the three strongest lines of the spectrum:  $X$ ,  $CX$ , and  $XX$ . We measured the PL spectra at different excitation powers. Figure 3 shows the power dependence of the integrated intensity of each of the three lines. The observed superlinear behavior of the  $XX$  line and the approximately linear one of

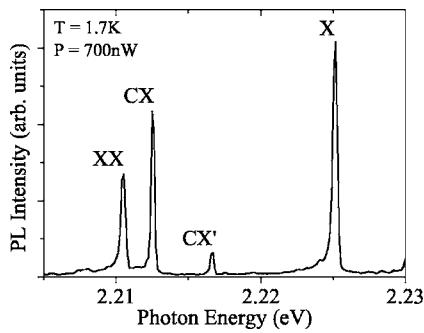


FIG. 2. Photoluminescence spectrum of the selected quantum dot, excited with a frequency-doubled pulsed beam of a Ti:sapphire laser at 402 nm (3 eV). The averaged excitation power was 700 nW.

X and CX lines support the assumed identification.

Final proof of this identification is provided by single photon correlation measurements presented below. The excellent mechanical stability of our experimental setup allowed us to follow the microphotoluminescence of this quantum dot during weeks of measurements. The main results of the paper were confirmed for several other quantum dots.

### C. Measurements of PL decay on individual QD

The time-dependent intensities of PL emission from X, CX, and XX states under pulsed excitation are presented in Fig. 4. Two components, a fast and a slow one, are present in all the recorded decays, in agreement with previous observations on CdTe/ZnTe QDs.<sup>1,2</sup> The fast component represents excitonic radiative lifetime, while the (much weaker) slow component, usually in II-VI systems attributed to dark exciton contribution,<sup>10,11</sup> is probably due to excitation delayed by some trapping processes.<sup>1</sup> The data was fitted with monoexponential decay applied to the “fast” part of the decay curve (as indicated in Fig. 4), producing radiative lifetime values  $\tau_X=290\pm 50$  ps,  $\tau_{CX}=330\pm 50$  ps,  $\tau_{XX}=190\pm 50$  ps, for exciton, charged exciton, and biexciton, respectively. The ratio of the exciton decay time to the biexciton decay time is equal to 1.5, which is consistent with previous results obtained on CdTe/ZnTe (Ref. 2) and InAs/GaAs (Ref. 12) QDs. The

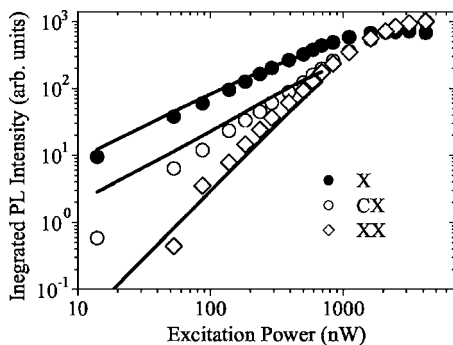


FIG. 3. Integrated photoluminescence intensity at  $T=1.7$  K as a function of excitation power for X, CX, and XX. Solid lines show model calculation (see Sec. IV B).

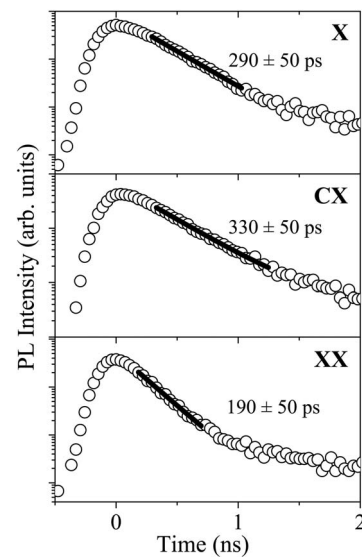


FIG. 4. Intensity of X, CX, and XX photoluminescence as a function of time following excitation pulse. Solid lines represent exponential decays with indicated lifetime values. Excitation power 700 nW, temperature 1.7 K.

decay time values obtained were used in the analysis of subsequent experiments.

### D. Excitation correlation spectroscopy

The ECS technique, used mainly for studies of quantum wells,<sup>13</sup> has previously been applied to QDs to study coherent processes.<sup>14,15</sup> It provides an excellent temporal resolution, limited principally by the width of the exciting laser pulse. We performed preliminary ECS experiments at non-resonant excitation to check the time scale of QD excitation processes, inaccessible in our single QD PL decay measurements. Figure 5 shows integrated intensity of the three PL lines from a selected QD as a function of the delay between the two excitation pulses. A modification of the signal is observed in the vicinity of zero delay, originating from superlinear (increase) or sublinear (decrease) excitation power dependence. Besides the effects on the scale of the radiative

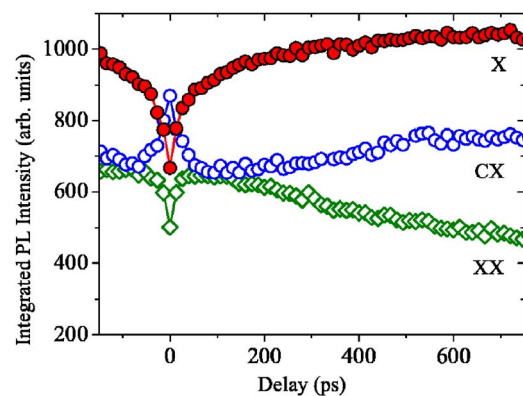


FIG. 5. (Color online) Integrated intensity of X, CX, and XX lines vs delay between two excitation pulses in ECS experiment. Excitation power of a single beam 230 nW, temperature 1.7 K.

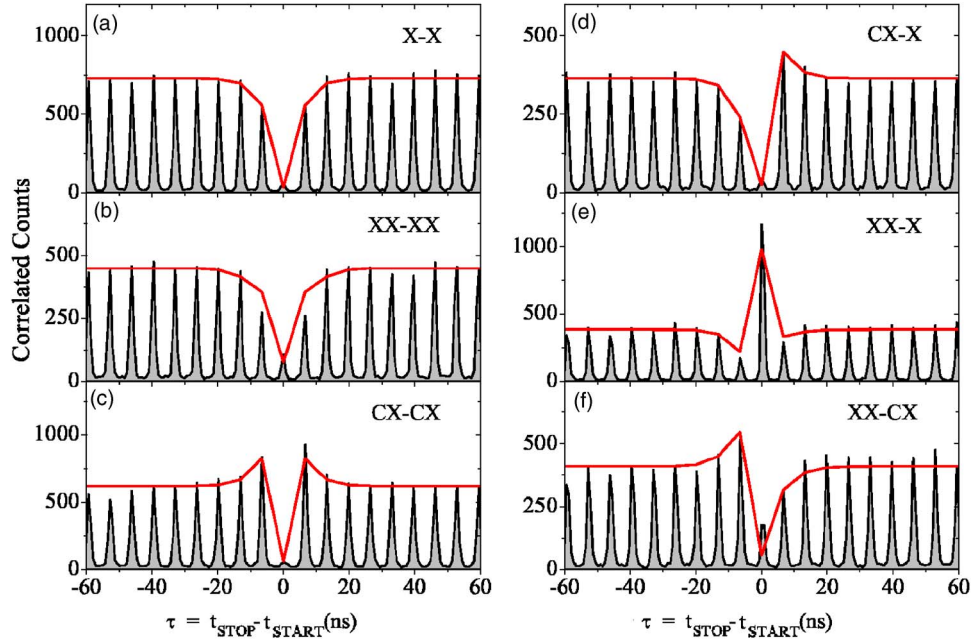


FIG. 6. (Color online) Histograms of correlated counts as a function of time interval  $\tau = t_{STOP} - t_{START}$  between photon registration events in start and stop detectors, tuned to indicated transitions (start-stop order). The average excitation power 700 nW, repetition period 6.6 ns, and time bin 733 ps. Acquisition time 1 h (a, d, e) or 2.5 h (b, c, f). Single count rates 9300/s, 5800/s, and 4600/s (first detector), 4500/s, and 3900/s (second detector) for X, CX, and XX, respectively. Solid lines represent fits of rate equation model (see Sec. IV B), calculated for each peak and joined with a line for better visibility.

recombination times (hundreds of ps), pronounced sharp structures are visible on a scale of order of tens of ps. We attribute them to trapping of carriers and excitons by the QD, in agreement with the discussion of time resolved measurements on the QD ensemble in Sec. II. A detailed discussion of ECS measurements will be presented elsewhere.

### E. Single photon correlation measurements

The results of correlation measurements involving X, CX, and XX states from the chosen QD are presented in Fig. 6. Each of the six histograms consists of distinct peaks separated by a 6.6 ns excitation repetition period. No background subtraction was made. The signal between the peaks is negligible, indicating that the emission from the QD is truly locked to the pulsed excitation. The strong suppression of the central peak at  $\tau=0$  (antibunching) visible in X-X, CX-CX, and XX-XX autocorrelation histograms [Figs. 6(a)–6(c)] constitutes evidence of single photon emission. An enhancement of the central peak in XX-X cross-correlation [Fig. 6(e)] confirms the presence of the biexciton-exciton cascade, while its suppression in cross-correlations between different QD charge state transitions [Figs. 6(d) and 6(f)] confirms our identification, as no X-CX or XX-CX cascades are expected. These results confirm the potential of CdTe/ZnTe QDs as sources of single photons or photons pairs “on demand,” in agreement with previous reports.<sup>2</sup>

Apart from the effects on the zero delay peak, all the histograms presented show that more than one excitation period is necessary to reach a steady state. Similar long time scale memory effects (blinking), observed previously in autocorrelation measurements on InAs/GaAs QDs, were inter-

preted recently<sup>3,16</sup> in terms of QD charge state variation. We present here a systematic study of these effects by measuring various types of correlations.

To check the evolution of the QD state between the excitation pulses we performed some of the correlation measurements at two different repetition periods  $T_{rep}=6.6$  or 13.2 ns. We established that the correlation functions did not depend on the time interval between excitation pulses, as seen in Fig. 7. In order to evaluate function  $g^{(2)}(n)$  integrated counts  $C(n)$  of the peak number  $n$  were normalized according to the for-

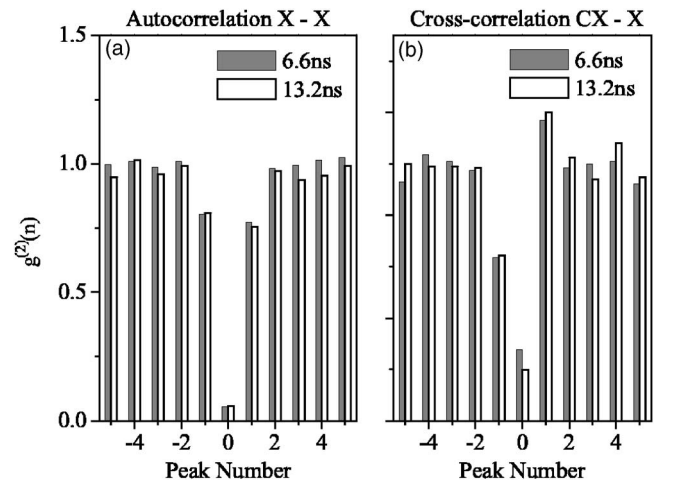


FIG. 7. X-X (a) and CX-X (b) second order correlation functions determined with two different repetition periods  $T_{rep}=6.6$  ns (gray bars) or 13.2 ns (white bars), plotted as a function of peak consecutive number  $n = \tau/T_{rep}$ .

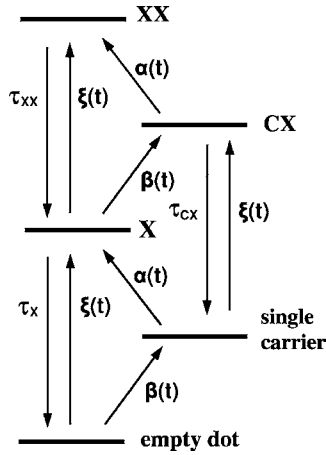


FIG. 8. Scheme of energy levels and optical transitions considered in the model.  $\alpha(t)$ ,  $\beta(t)$ , and  $\xi(t)$  are the time dependent capture rates of oppositely charged single carriers and exciton, respectively. Constants  $\tau_x$ ,  $\tau_{cx}$ , and  $\tau_{xx}$  are decay times of respective states.

mula:  $g^{(2)}(n) = C(n)/(N_1 N_2 T_a T_{rep})$ , where  $N_{1,2}$  are single counters rates,  $T_a$  is total acquisition time, and  $T_{rep}$  is repetition period.<sup>17</sup> The negligible contribution of background counts was not taken into account in the calculation.

In other words the peak consecutive number  $n = \tau/T_{rep}$  is a good parameter to present our correlation results, rather than the time coordinate used for other systems.<sup>16</sup> This indicates that the QD state remains frozen between the excitation pulses within the accuracy of our correlation experiment, no matter whether the correlated transitions occur in the same [Fig. 7(a)] or in different [Fig. 7(b)] QD charge states. Therefore in a further discussion we shall ignore in particular the long time-scale component observed in the PL decay measurements.

#### F. Capture of single carriers by the QD

The results of  $CX$ - $X$  correlation measurements provide direct evidence for the important role of single carrier capture in QD excitation. Indeed,  $CX$ - $X$  coincidences are registered only if the charge state of the dot changes between the correlated emission events. If the carriers were injected predominantly in pairs possessing no effective charge, changes in the charge state of the dot would be unlikely, resulting in  $CX$ - $X$  coincidence rates much smaller than  $X$ - $X$  or  $CX$ - $CX$  autocorrelation count rates, in contrast with our experimental results. Another argument for single carrier excitation comes from the strong asymmetry of the  $CX$ - $X$  and  $XX$ - $CX$  correlation histograms (a similar result has been obtained by Kiraz *et al.*<sup>18</sup> on InAs/GaAs QDs and Chang *et al.*<sup>19</sup> on InGaAs/GaAs QDs with cw excitation). Peaks at positive (negative) time delay in a  $CX$ - $X$  histogram [Fig. 6(d)] represent detection of a neutral exciton photon after (before) charged exciton photon. In particular reexcitation of the quantum dot directly after the charged exciton recombination ( $n=1$  peak) requires a single carrier to be trapped, while three carriers are necessary for the opposite emission order ( $n=-1$ ). The much smaller probability of three trapping

events following a single pulse results in smaller intensity of the  $n=-1$  peak than that of the  $n=1$  one. After several pulses the QD approaches a steady state with nonzero probabilities of finding the dot in a neutral or charged state, resulting in an intermediate asymptotic peak intensity. A similar explanation holds for the  $XX$ - $CX$  cross-correlation [Fig. 6(f)].

As reported previously in respect to  $X$  emission from non-resonantly excited III-V QDs,<sup>3</sup> we also observed the antibunching of photons emitted in  $n \neq 0$  pulses: lower than average intensities of peaks at both sides of the central one [Fig. 6(a)]. This indicates that excitation to  $X$  state is less probable when the dot is empty (following an  $X$  photon emission) than when it is in steady state condition. This difference between the steady state and empty ground state of the dot can be naturally explained in terms of the QD charge. In the case of  $XX$  autocorrelation [Fig. 6(b)] the decrease of  $n = \pm 1$  peaks is even more pronounced. This is because as many as four carriers are needed to repopulate the  $XX$  state. In the steady state the finite probability of the presence of a carrier in the quantum dot decreases the average number of carriers necessary to repopulate the radiative state in both cases ( $X$  or  $XX$ ). An explanation of the suppression of  $n = \pm 1$  peaks in  $XX$ - $X$  cross-correlation histogram [Fig. 6(e)] may be obtained in the same spirit.

The opposite conduct is observed in the charged exciton autocorrelation:  $n = \pm 1$  peaks are higher than the  $n=0$  one [Fig. 6(c)]. This result is explained by the fact that after  $CX$  recombination the dot contains a single carrier, and two more carriers (of opposite sign) must be captured to repopulate the  $CX$  state. Peaks far from  $n=0$  are less intense because the steady state involves nonzero probability of a neutral empty QD state, requiring three carriers to enable the QD to emit another  $CX$  photon.

In summary, we explain the observed memory effects in terms of the QD charge state variation caused by capture of single carriers by the QD. Within our interpretation we would not expect any memory effects without single carrier capture: all  $n \neq 0$  peaks would have the same intensity (since  $\tau_x \ll T_{rep}$ ). The above analysis allows us to make qualitative predictions concerning correlation experiments performed under continuous wave excitation. The suppression of  $X$ - $X$  and  $XX$ - $XX$  autocorrelation in the neighborhood of zero delay should produce a broadening of the antibunching dip in the case of cw excitation. The same suppression observed in  $XX$ - $X$  cross-correlation should lead to a narrowing of the bunching peak in cw experiments. Therefore the contribution of single carrier capture can help to explain the unexpectedly wide antibunching dip in  $X$ - $X$  autocorrelation under cw excitation reported for CdTe/ZnTe QDs,<sup>2</sup> while no broadening was observed for the  $XX$ - $X$  cross-correlation peak. Furthermore, our results indicate that lifetime determination of QD excitons based on an analysis of cw correlation measurements<sup>20,21</sup> may lead to significant errors if single carrier capture is neglected. In other words, the recovery time measured in the correlation experiments should not be confounded with the spontaneous emission lifetime, which can be observed in PL decay.

## IV. MODEL DESCRIPTION OF THE EXPERIMENTAL RESULTS

### A. Rate equation model

A simple model was introduced to describe our results quantitatively. It includes occupation probabilities of five QD states (Fig. 8), presented as a vector  $\mathbf{n}(t) = [p_0, p_c, p_X, p_{CX}, p_{XX}]$  where  $p_i$  is the probability of finding the  $i$ th QD state occupied at time  $t$ . Since the QD confining potential is shallow,<sup>22</sup> we do not include neutral or charged excitonic levels above the biexcitonic one. Due to the low transition intensity of one of the charged excitons in the PL

spectrum we neglect charge states of one type. Then, possible transitions related to the capture of a carrier or of a neutral exciton as well as to radiative recombination are included as presented in Fig. 8.

Since the model deals only with level occupations, it cannot describe any coherent effects such as, e.g., polarization conversion.<sup>23</sup> It also neglects the fine structure of the excitonic states (e.g., dark excitons). Within the assumed approximations, the five occupation probabilities sum to unity. The dynamics of the occupation vector can be expressed by the equation:  $d\mathbf{n}/dt = A(t)\mathbf{n}(t)$  where  $A(t)$  is a transition rate matrix describing radiative decays and excitation processes:

$$A(t) = \begin{bmatrix} -\alpha(t) - \xi(t) & 0 & 1/\tau_X & 0 & 0 \\ \alpha(t) & -\beta(t) - \xi(t) & 0 & 1/\tau_{CX} & 0 \\ \xi(t) & \beta(t) & -\alpha(t) - \xi(t) - 1/\tau_X & 0 & 1/\tau_{XX} \\ 0 & \xi(t) & \alpha(t) & -\beta(t) - 1/\tau_{CX} & 0 \\ 0 & 0 & \xi(t) & \beta(t) & -1/\tau_{XX} \end{bmatrix}. \quad (1)$$

The important role of trapping of single carriers has already been discussed above. We also include the possibility of excitation by trapping free excitons which (as follows from our simulations) is needed to achieve the observed quadratic excitation-power dependence of  $XX$  line intensity. The characteristic time scales of the electron, hole, and exciton trapping (which may be different from one another) are larger than the laser pulse width because of relaxation processes within the barrier material. With the small outer barrier thickness of 50 nm we expect the trapping rates to rise almost instantaneously and to decay with characteristic times of the order of the barrier exciton lifetime (Sec. II). Therefore we assumed as a starting point a common exponential time dependence of the three excitation rates, with a decay time of  $\tau_{exc} = 20$  ps. As we know the three radiative lifetimes  $\tau_X$ ,  $\tau_{CX}$ , and  $\tau_{XX}$  from independent experiments, the only free parameters of the model remain constants  $\alpha$ ,  $\beta$ , and  $\xi$  understood as time-integrated respective capture rates, e.g.,  $\alpha(t) = (\alpha/\tau_{exc})\exp(-t/\tau_{exc})\theta(t)$ , where the Heaviside function  $\theta(t)$  equals 0 for  $t < 0$  and 1 for  $t \geq 0$ . We also allow for an arbitrary coefficient between the computed and measured count rates to account for (unknown) photon detection efficiency.

We integrated the rate equations numerically to compute  $\mathbf{n}(t)$ . Integration over time of  $I(t) = p_X/\tau_X, p_{CX}/\tau_{CX}, p_{XX}/\tau_{XX}$  gives PL intensities (per excitation pulse) for  $X$ ,  $CX$ , or  $XX$ , respectively. The initial state is defined by a steady state or by a defined transition observed in the correlation experiment.

If we choose the initial point for integration to meet the steady state condition  $\mathbf{n}(t) = \mathbf{n}(t + T_{rep})$ , we get relative line intensities in the PL spectrum. As far as the correlation experiment is concerned, the first photon unequivocally defines  $\mathbf{n}(0)$  (e.g., if we detect a photon from the recombination of  $CX$ , we know the dot is occupied by respective single car-

rier) and therefore we can calculate the expected PL intensity for the following  $T_{rep}$  period. The correlation function is proportional to this expected PL multiplied by the average PL intensity of the line of the first photon. The calculation of the correlation function  $g^{(2)}(\tau)$  for the central peak is slightly more complicated and can be written as

$$g^{(2)}(\tau=0) = \frac{1}{I_{AB}} \int_0^{T_{rep}} \int_{\tau'}^{T_{rep}} [I_{A,s}(\tau') I_{B,a}(\tau'') + I_{B,s}(\tau') I_{A,b}(\tau'')] d\tau' d\tau'', \quad (2a)$$

$$I_{AB} = \int_0^{T_{rep}} I_{A,s}(\tau') d\tau' \int_0^{T_{rep}} I_{B,s}(\tau'') d\tau'', \quad (2b)$$

where in the example case of  $X$ - $CX$  cross-correlation  $A$  denotes the  $X$  state,  $a$  denotes the QD state after  $X$  photon emission (empty dot),  $B$  denotes the  $CX$  state,  $b$  denotes the QD state after  $CX$  photon emission (a respective single carrier present), and  $s$  denotes the steady state.  $I_{K,m}(t)$  is time dependence for a rate of emission from a QD state denoted as  $K$  when starting from the state denoted as  $m$ .

### B. Description of the experimental results

The results of fitting the rate equation model to experimental data are presented in Figs. 3 and 6. The fitting procedure takes into account the results of PL intensity power dependence and the complete set of auto- and cross-correlation measurements simultaneously. As seen in Fig. 6, the model describes the results of correlation measurements with excellent accuracy. The shapes of the correlation histograms, i.e., long time-scale bunching and antibunching fea-

tures, are repeated by the model with high fidelity. The calculated number of counts in the zero delay peak on each histogram in Fig. 6 agrees with values obtained in experiment. The same set of fitting parameters allows the slopes and magnitude of  $X$ ,  $CX$ , and  $XX$  emission intensities to be described, plotted as a function of excitation power in Fig. 3. However, the region of strong, saturating excitation powers is not described adequately by the model (not shown). This aspect requires further study.

As a result of the fitting procedure, integrated capture rates per pulse for single carriers and exciton were obtained:  $\alpha=0.80$ ,  $\beta=0.86$ , and  $\xi=0.26$ , respectively. We see that the capture of a single carrier is more than three times more probable than capture of an electron-hole pair. This supports our interpretation (Sec. III F) of the results of the auto- and cross-correlation experiment indicating the role of single carrier capture processes in QD excitation.

The systematic simulations have convinced us that the integrated capture rates are the most important parameters influencing the PL intensity and correlation results. The detailed temporal distribution of the capture processes is less important, provided that their characteristic times are significantly lower than the QD PL decay times or the excitation

repetition period. To study these processes in detail, different tools should be applied, such as pump-probe absorption measurements or correlation excitation spectroscopy.

## V. CONCLUSIONS

We demonstrate the utility of photon correlation measurements to study nonresonant excitation mechanisms of semiconductor quantum dots. A qualitative analysis indicates the important role of single carrier capture processes and leads to an explanation of the unusually wide antibunching dip observed in previously reported autocorrelation measurements under cw excitation. The rate equation model introduced allowed us to describe a complete set of correlation and PL intensity results and to obtain quantitative information on the trapping rates of single carriers and excitons by the quantum dot.

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