Blinking Statistics and Excitation-Dependent Luminescence Yield in Si and CdSe Nanocrystals

Benjamin Bruhn, Fatjon Qejvanaj, Ilya Sychugov and Jan Linnros*

Materials and Nanophysics Department, ICT School, KTH - Royal Institute of Technology,
16440 Kista, Sweden

**corresponding author: ilyas@kth.se*

Keywords: quantum dots, photoluminescence, intermittence, silicon, cadmium selenide

Abstract

Blinking is a phenomenon observed in single quantum emitters, which reduces their overall light emission. Even though it seems to be a fundamental property of quantum dots (QDs), substantial differences can be found in the blinking statistics of different nanocrystals. This work compares the blinking of numerous single, oxide-capped Si nanocrystals with that of CdSe/ZnS core-shell nanocrystals, measured under the same conditions in the same experimental system and over a broad range of excitation power densities. We find that ON- and OFF-times can be described by exponential statistics in Si QDs, as opposed to power-law statistics for the CdSe nanocrystals. The type of blinking (power-law or mono-exponential) does not depend on excitation, but seems to be an intrinsic property of the material system. Upon increasing excitation power, the duty

cycle of Si quantum dots remains constant, whereas it decreases for CdSe nanocrystals, which is readily explained by blinking statistics. Both ON-OFF and OFF-ON transitions can be regarded as light-induced in Si/SiO₂ QDs, while the OFF-ON transition in CdSe/ZnS nanocrystals is not stimulated by photons. The differences in blinking behavior in these systems will be discussed.

Introduction

An inherent problem of fluorophore molecules and quantum dots in terms of light emission efficiency is so-called blinking, a random cyclic transition between a bright (ON) and a dark (OFF) state. For quantum dots it has first been observed in CdSe nanocrystals^{1,2} and since then has also been confirmed for other semiconductor materials, including silicon.³ A quantum dot in the OFF state absorbs energy, but, instead of emitting photons, loses the acquired energy to lattice vibrations, possibly due to Auger process, which reduces its quantum efficiency.

In general, quantum dots are recognized as advantageous compared to organic molecules regarding stability against photobleaching, where irreversible degradation can make the molecule permanently dark.⁴ However, quantum dots can exhibit reversible changes of the luminescence yield as a function of excitation power, which are examined here in detail on a single particle level for two different material systems: CdSe/ZnS and Si/SiO₂.

There are many models attempting to explain blinking in a general way. The earliest models described the transition from the bright to the dark state as a two-step ionization process: One charge carrier of an exciton is ejected from the quantum dot, leaving behind a charged core. Subsequently, additional excitons recombine via non-radiative Auger processes.^{2, 5} Thermal contributions to the ionization were proposed, as well.⁶ A large portion of the literature on blinking finds power-law distributions of ON- and OFF-times, in particular for the frequently used model system CdSe/ZnS. A random walk in phase-space with transitions between intrinsic excited state and a trap state^{7, 8}, as well as a stationary exponential distribution of trap levels in terms of distance from the quantum dot⁹ were proposed to explain this behavior. A different approach is based on the assumption that temporary non-radiative channels in or close to the quantum dot are responsible for the dark state.^{10, 11} There is still a heavy debate on which of the

two classes of models is correct, or even a combination of them. Another important question concerns the role of four-particle Auger recombination (due to simultaneous excitation of two excitons in the same nanocrystal) as the main cause for ON-OFF transitions, as opposed to a single-photon absorption mechanism or thermal processes.

We have found exponential distributions for single oxide-capped silicon nanocrystals,¹² apparently contradicting previous results on porous silicon particles under high excitation,¹³ where power law statistics had been observed. What causes this difference is an important question that needs to be addressed. Most studies of the CdSe/ZnS system are also performed at high excitation (kW/cm^2), and it could not be excluded that blinking statistics might be dependent on excitation power. Therefore we decided to conduct an investigation at low excitation conditions for CdSe nanocrystals to clarify whether blinking statistics changes between different excitation regimes.

This work comprises a comparative study of the two nanocrystal systems Si/SiO₂ and CdSe/ZnS in the same experimental setup and at varying excitation power densities, spanning a range of two orders of magnitude. It shall be clarified whether the blinking statistics can be altered by excitation power itself or if it can be understood as an intrinsic property. The question of light-induced transitions between ON and OFF state will be tackled and, additionally, the dependency of the quantum yield on excitation power density will be discussed.

Experimental

The silicon sample was fabricated from a plain, low doped ($20 \text{ } \Omega\cdot\text{cm}$) 10 mm X 10 mm Si (110) wafer piece, using electron-beam lithography, reactive ion etching and oxidation.¹⁴ First, negative resist (HSQ) was spun onto the cleaned sample at 6000 rpm and baked at 150 °C for 10

min, then the resist was exposed and subsequently developed in MF-CD26 for 1 min. A 40 s long plasma etching step using HBr chemistry transferred the mask pattern into the silicon, resulting in approximately 300 nm high silicon walls. Oxidation in the self-limiting regime¹⁵ at 900 °C for 5 h then reduced the core size of the silicon structures until spatially well separated, luminescing silicon nanocrystals were obtained.

The CdSe sample was prepared by spinning commercially available CdSe/ZnS quantum dots (Evident Technologies EviDotsEB-C11-TDL-0600) in a strongly diluted solution onto a clean glass cover slide at 10000 rpm. These CdSe quantum dots have a solid-state shell of ZnS and are free of organic ligands. The thickness of the ZnS shell was estimated to be ~ 3 monolayers from combined AFM and PL measurements. The nanocrystals are of spherical shape as seen in high resolution SEM micrographs (not shown).

Photoluminescence images of both the silicon and cadmium selenide sample are shown in Figure 1. The first contains ordered arrays of (mostly) single Si quantum dots, whereas the latter is covered with random patterns of single and clustered CdSe nanocrystals.

PL measurements were carried out using an Omicron Phoxx 405 nm diode laser for excitation and an inverted Zeiss Observer.Z1m microscope with a 100x objective lens (0.9 NA for CdSe and 0.7 NA for Si), equipped with a thermoelectrically cooled Andor iXon3 888 EMCCD camera, for detection. While the CdSe sample was excited through the objective lens (bright field) at excitation power densities of 6, 13, 63, 125 and 313 W/cm², dark field excitation (laser beam incident at an angle of approximately 50 degrees) at 8, 40, 80, 120, 160 and 200 W/cm² was used for the silicon sample. Image sequences of 20000 frames (with 100 ms - 1 s acquisition time at 8, 40 and 80 W/cm² and 100 ms at 80, 120, 160 and 200 W/cm² excitation power density) and 54000 frames (with 10 ms – 100 ms acquisition time at 6, 13 and 63 W/cm²

and 100 ms at 63, 125 and 313 W/cm² excitation power density) were recorded for Si and CdSe, respectively. The readout time was less than 20 percent and 2 percent for the 10 ms and 100 ms frame acquisition time for cadmium selenide and silicon, respectively.

Analysis of the image sequences was carried out using the same in-house-built ImageJ plugins as in the previous work.¹² The quantum dots were mapped and their intensity was extracted from the different blinking sequences. A threshold was applied to transform the resulting blinking traces into binary (ON/OFF) traces for use in the statistical analysis.

Results

It is believed that if a blinking trace exhibits two distinct levels, a single quantum emitter is being observed. In this work, numerous Si dots with such traces could be found, but only a few CdSe dots. It has been reported¹⁶ that CdSe nanocrystals generally blink on shorter time scales than silicon, which can cause ON and OFF levels to be poorly resolved for long time bins, as transitions between the two states during the acquisition of an image smear out the extracted intensity levels.

Figure 2 shows typical blinking traces of single quantum dots at different excitation powers. At low excitation power, the CdSe nanocrystal clearly alternates between two states, while at higher excitation its ON-level is not as well defined anymore. This can readily be explained by the shortening of ON and OFF intervals below the temporal resolution limit of the measurement setup. The SiQD, on the other hand, exhibits significantly longer intervals; however, its switching frequency also increases with increasing excitation.

The emission intensity of a nanocrystal can be extracted from the blinking trace as the difference between the ON-level intensity and the OFF-level intensity, where the latter

represents the local background, which usually increases linearly with excitation power. Different nanocrystals can emit with largely varying amplitudes, which can be traced back to differing ratios of radiative and non-radiative exciton recombination rates, i.e. quantum efficiencies. Below an average occupation of one exciton per nanocrystal, the amplitude increases linearly, while above the threshold usually a sub-linear increase can be observed, caused by competing Auger-recombination when several excitons are present.¹⁷ In Figure 2, the silicon nanocrystal has seemingly entered the several-excitons regime at high excitation, as the intensity is not increasing at the same rate as for low pumping. Due to drastically shortened ON intervals in the CdSe quantum dot the ON-level cannot be resolved properly anymore, which poses a problem for the determination of the blinking amplitude. That a single dot is being observed is not apparent from the high excitation blinking trace, but only from the fact that the same nanocrystal is being probed that exhibits clear two-level blinking at lower excitation. However, the focus of this work does not lie on the blinking amplitude, but rather on the blinking statistics of Si and CdSe quantum dots and a comparison of the two systems. Finally we note that the duty cycle (fraction of time spent in ON state) decreases for the CdSe dot, but slightly increases for the silicon one.

In order to extract data regarding the blinking statistics, every trace is first being transformed into a binary trace (ON, OFF) by applying a threshold. All intensity values above the threshold are translated into ON and all below it to OFF. Two separate lists are then formed, one containing the lengths of all ON intervals and the other one containing all OFF intervals. The duty cycle can easily be obtained by dividing the total ON time by the sum of total ON and total OFF time. A histogram of the ON and OFF interval lengths is referred to as an ON- or OFF-time distribution. To account for the insufficient experimental time, i.e. to introduce higher statistical

accuracy for long intervals, the counts in those histograms are recalculated into probability densities.¹⁸

Figure 3 displays such ON- and OFF-time distributions of single quantum dots for different excitation powers. As is also frequently found in the literature^{7, 19} CdSe nanoparticles obey pure power-law blinking for the OFF-times, i.e. OFF-ON transitions (Figures 3a, 3b). A truncated power-law with excitation-dependent truncation can be observed for ON-OFF transitions. Shortening of the truncation time for increasing excitation has been reported before as well.¹⁹ The power-law exponent is usually found in the vicinity of $m = 1.5$. Silicon quantum dots, on the other hand, exhibit mono-exponential ON- and OFF-time distributions (Figures 3c, 3d). Altogether we looked at about 900 traces of Si dots and selected about 30 objects with distinct ON and OFF states and moderate switching frequency for analysis. It should be noted that among them 3 dots with apparent (truncated) power-law distributions were observed, but omitted as outliers. A more thorough investigation of these could be the subject of future work, as in other work¹³ a large number of such nanocrystals have been reported. However, several emitters could simply be present in the same optically resolved spot, since a superposition of several quantum dots with exponential statistics and different blinking frequencies would lead to an apparent power-law distribution. Such issues can often be avoided by only selecting traces with two clear, well separated levels for analysis (see e.g. Figure 2b).

An average blinking frequency is defined when the interval lengths are distributed exponentially, namely as τ in $p=A*\exp(-t/\tau)$. It can also roughly be determined by dividing the number of blinking events by the total experimental time. When the ON and OFF intervals follow a power-law distribution, which is inherently scale-invariant, an average blinking frequency does not exist. However, in a truncated power-law, the exponential cutoff may serve

as a measure for an average frequency. Figure 4 shows the ON-OFF and OFF-ON transition frequencies for a silicon quantum dot as a function of excitation power. The statistical plot contains data from numerous single nanocrystals, showing that on average both ON- and OFF-blinking frequencies increase linearly with excitation. Due to the above mentioned reasons a similar plot for CdSe QDs cannot be shown. It may be noted, however, that the ON intervals become increasingly shorter for higher excitation in that system.

Discussion

Mono-exponential interval length distributions can be interpreted in two ways. In the charge model class picture, a single trap state in the vicinity of the quantum dot can capture the electron or hole of an exciton, so that the remaining carrier enables non-radiative three-particle Auger processes in the quantum dot core.⁵ Another possibility is given in the enhanced non-radiative channel picture, where a temporary trap in or close to the quantum dot allows excitons to recombine without the emission of a photon.¹⁰ A return to the bright state occurs through neutralization, i.e. tunneling of the trapped carrier back into the core and recombination with the opposite carrier residing there, or closing of the fast non-radiative channel, respectively. On the other hand, power-law distributions can originate from more complicated systems, for which a wide variety of models is available, as described in the Introduction.

Our results confirm the findings of most works for CdSe/ZnS quantum dots, namely pure power-law distributions for OFF-time interval lengths and truncated power-law distributions for ON-time intervals even at low excitation. The exponential tail is found to be excitation intensity dependent for ON-time interval lengths. On the other hand, in the case of well passivated silicon nanocrystals, mono-exponential distributions were found for both ON- and OFF-times,

confirming previous experiments.¹² This is different from porous silicon particles,¹³ where high excitation power densities and relatively thin oxide shells were used. Thus in the current work it has been established that the excitation power density does not change the blinking statistics between mono-exponential and power-law type of behavior.

Figure 4 demonstrates that both ON-OFF and OFF-ON transition rates in silicon nanocrystals are linearly dependent on the excitation power, indicating that both processes are photoinduced. Since the Auger process relies on co-existence of two excitons in a nanocrystal and the probability of the two-exciton formation increases quadratically with the excitation power¹¹ this effect can be ruled out in the investigated excitation power regime. Instead, it was shown that photoionization rate for a quantum dot can indeed grow linearly with the excitation power.^{20, 21} Together with the mono-exponential ON- and OFF-time distributions, simple opening and closing of an efficient non-radiative recombination center by a single photon within or close to the quantum dot can provide a reasonable explanation.

In CdSe nanocrystals, ON and OFF intervals do not follow an exponential distribution, but a power-law, which is scale-invariant and therefore does not have any characteristic blinking frequency. The OFF-ON transition is independent of excitation power, as Figure 3a shows. This fact points towards a process that solely relies on e.g. tunneling of a trapped charge carrier back into the quantum dot core for neutralization. Transitions from the ON to the OFF state are also distributed according to a power-law, however, an exponential tail can be observed at long times (in the order of seconds), which is excitation dependent and therefore hints at a process induced by photon absorption. There is an ongoing discussion about the excitation power dependence of the exponential cutoff, namely whether the inverse truncation time increases linearly or quadratically with power.²² Both the occurrence of power-law statistics and very frequent

transitions between ON and OFF in CdSe quantum dots suggest a significantly higher availability of dark states and therefore a considerably increased switching probability. Also note that the truncation time in ON-OFF blinking strongly depends on the environment.²³ It is believed that the quality of surface passivation, and therefore the existence of surface states, plays a major role in blinking.²⁴

Now we turn to a point, which is of major importance for applications and pump-dependent experiments, namely the dependence of the duty cycle of blinking quantum dots on the excitation power. The detailed discussion on this topic is lacking in the literature, although it was noticed previously that such dependence actually takes place for CdSe nanocrystals.²⁵

For the case of silicon nanocrystals the duty cycle D (the ratio of time spent in ON-state to the total experimental time) can be defined by the number of time bins N spent in a corresponding state:

$$D \equiv \frac{N_{ON}}{N_{ON} + N_{OFF}} = \frac{\sum_{n=1}^{\infty} A_{on} \cdot \exp(-n / \tau_{on}) \cdot n}{\sum_{n=1}^{\infty} A_{on} \cdot \exp(-n / \tau_{on}) \cdot n + \sum_{n=1}^{\infty} A_{off} \cdot \exp(-n / \tau_{off}) \cdot n} \quad (1).$$

Taking into account the fact that the total number of ON-OFF switching events equals to the number of OFF intervals (and vice versa):

$$\sum_{n=1}^{\infty} A_{on} \cdot \exp(-n / \tau_{on}) = \sum_{n=1}^{\infty} A_{off} \cdot \exp(-n / \tau_{off}) \quad (2),$$

and, using series sum formulas and Taylor expansion, equation (1) becomes

$$D \approx \frac{1}{\frac{1}{\tau_{off}} + \frac{1}{\tau_{on}}} \equiv \frac{f_{01}}{f_{01} + f_{10}} \quad (3).$$

As both OFF-ON and ON-OFF switching frequencies f_{01} and f_{10} increase linearly with the excitation power (cf. Figure 4b) the duty cycle remains constant (shown as a straight line in Figure 5).

For CdSe and most other material systems the situation is different. ON-OFF transition rates are distributed according to a truncated power law with excitation-dependent exponential cutoff. OFF-ON switching events follow a pure power-law. Thus, the number of time bins in ON- and OFF-states is described by following sums:

$$N_{\text{ON}} = \sum_{n=1}^{\infty} A_{\text{on}} \cdot n^{-m} \exp(-n / \tau_{\text{on}}) \quad (4),$$

$$N_{\text{OFF}} = \sum_{n=1}^{\infty} A_{\text{off}} \cdot n^{-m} \quad (5).$$

The duty cycle was in this case calculated numerically and is shown as a function of excitation power in Figure 5, where it is seen reducing with increasing excitation power (also apparent from the time traces displayed in Figure 2a). For both ON- and OFF-time power laws an exponent of $m = 1.5$ has been chosen, which is a widely accepted value found in literature and in this work (cf. Figure 3). In order to place data points from measured quantum dots in Figure 5 the data were normalized to account for dot-to-dot variations in, e.g. blinking amplitude. The extrapolated intersection of an individual curve with the abscissa was defined as unity, while for the excitation power the value where the intensity reduces to 20 % was set to unity for a given nanocrystal. No error bars are given since a binary ON-OFF trace has been analyzed here.

In general, with increasing excitation power the number of incident photons increases linearly. A single-photon absorption related mechanism causes a state transition from bright to dark in quantum dots. Therefore, a linear dependency of the inverse truncation time for CdSe nanocrystals in the ON-time power law distribution, also referred to as the exponential cutoff

parameter, on excitation power density is observed. This assumption appears in different works in the literature.^{5, 8} The return of the quantum dot to its emissive state cannot be mediated by photons and is therefore not dependent on excitation power, resulting in a pure power-law distribution of OFF-interval lengths. It is seen in Figure 5 that there is a significant decrease of the duty cycle upon increasing excitation power for CdSe nanocrystals, related to their power law blinking distribution, while silicon nanocrystals with exponential blinking statistics are robust against pump-induced changes in quantum yield. The argument for a linear dependency of the inverse truncation time in the ON-time distribution for CdSe nanostructures could be reinforced by simulation results shown in the figure.

As a unified physical model of the observed behavior we tentatively propose the following scenario. In Si/SiO₂ system trap sites located far away from the nanocrystal core and only photon-assisted processes can facilitate charge capture and release. Although a distribution of trap sites is possible its presence is not detected since the rate limiting process is photon absorption. On the other hand, in CdSe/ZnS system traps are easily accessible for carrier tunneling and their distribution reveals itself in the observed power-law. The trap sites are close to the nanocrystal core and direct tunneling is then responsible for more frequent blinking in CdSe nanoparticles. In the ON-state the photon absorption process competes with tunneling effect, but sets in only for long ON-times as power-law dominates short-time behavior, leading to the observed truncation of the power-law at shorter times with higher pumping. Finally in some Si QDs traps, as an exemption, trap sites can be close to the core yielding deviations from the exponential blinking statistics. The important effect of surrounding matrix and defect density on the blinking statistics will be investigated in a separate study.

Conclusions

To conclude, the blinking type (power law or exponential) can be regarded as an intrinsic property of the material system. In this case Si/SiO₂ nanocrystals in air exhibit mono-exponential distributions of ON and OFF times and CdSe/ZnS dots in air obey (truncated) power-law statistics. In CdSe the OFF-ON transition is independent of excitation while ON-OFF transitions can also be mediated by photon absorption. In Si both transitions are linearly dependent on excitation power, which hints at a single photon absorption process in both ways. Resulting from these differences, the duty cycle develops differently with excitation. In Si, it stays constant even for hard pumping, whereas it significantly decreases in CdSe. For light conversion applications, such as phosphors, oxide-passivated silicon quantum dots are not only appealing due to their non-toxicity and element abundance, but also because blinking does not lead to reduced luminescence quantum yield at high excitation.

Acknowledgements

Financial support from the Swedish Research Council (V.R.) through an individual contract and through a Linné grant is gratefully acknowledged.

Figures

Figure 1. PL images of the (a) silicon and (b) CdSe sample. The inset in (a) shows the part surrounded by the white square in higher magnification. Obviously, the silicon nanocrystals are arranged in a pattern defined by lithography, whereas the cadmium selenide quantum dots are distributed randomly after spincoating.

Figure 2. Blinking traces and corresponding intensity histograms of a single (a) CdSe and (b) Si quantum dot for different excitation power densities. An arbitrarily chosen threshold for forming a binary trace is indicated by a grey line.

Figure 3. (a, b) ON- and OFF-time distributions of a single CdSe nanocrystal. (a) A truncated power-law describes the experimental data well. The truncation time scales inversely with the excitation power. (b) Straight lines represent a pure power-law with exponent m , where m decreases for increasing excitation. (c, d) ON- and OFF-time distributions of a single Si nanocrystal; both are mono-exponential with decay time τ obtained from least squares fitting. All distributions have been shifted vertically for clarity.

Figure 4. (a) ON-OFF (squares) and OFF-ON (circles) blinking frequency of a single SiQD as a function of excitation power density showing linear increase. Straight lines are guides for the eye with $m = 1.0$. (b) Values of the slope m in a log-log scale for a large number of different single nanocrystals. The average value is unity. Significant noise in the extracted slope values leads to a spread in the data.

Figure 5. Duty cycle at different excitation power density values for a few single Si (light grey) and CdSe (black) nanocrystals in normalized units (see text). Most SiQDs do not change, while the two outliers are shown in color as empty squares (blue: SiQD from Figure 2, green: SiQD from Figure 3). A dashed line shows the constant duty cycle of the SiQDs deduced from their exponential statistics. On the other hand, a clear decrease in the ON fraction is observed for CdSe quantum dots with increasing excitation. The solid line is a result of a theoretical model where a pure power-law is assumed for OFF-time distributions and truncated power-law for ON-time with the truncation time scaling inversely with the excitation power.

References

- [1] Nirmal, M.; Dabbousi, B. O.; Bawendi, M. G.; Macklin, J. J.; Trautman, J. K.; Harris T. D.; Brus, L. E. Fluorescence Intermittency in Single Cadmium Selenide Nanocrystals. *Nature* **1996**, 383, 802-804.
- [2] Empedocles, S. A.; Norris, D. J.; Bawendi, M. G. Photoluminescence Spectroscopy of Single CdSe Nanocrystallite Quantum Dots. *Phys. Rev. Lett.* **1996**, 77, 3873-3876.
- [3] Mason, M. D.; Credo, G. M.; Weston, K. D.; Buratto, S. K. Luminescence of Individual Porous Si Chromophores. *Phys. Rev. Lett.* **1998**, 80, 5405-5408.
- [4] Resch-Gender, U.; Grabolle, M.; Cavaliere-Jaricot, S.; Nitschke, R.; Nann, T. Quantum Dots versus Organic Dyes as Fluorescent Labels. *Nature Meth.* **2008**, 5, 763-775.
- [5] Efros, Al. L.; Rosen, M. Random Telegraph Signal in the Photoluminescence Intensity of a Single Quantum Dot. *Phys. Rev. Lett.* **1997**, 78, 1110-1113.
- [6] Banin, U.; Bruches, M.; Alivisatos, P. M.; Ha, T.; Weiss S.; Chemla, D. S. Evidence for a thermal contribution to emission intermittency in single CdSe/CdS core/shell nanocrystals. *J. Chem. Phys.* **1999**, 110, 1195-1201.
- [7] Shimizu, K. T.; Neuhauser, R. G.; Leatherdale, C. A.; Empedocles, S. A.; Woo, W. K.; Bawendi, M. G. Blinking Statistics in Single Semiconductor Nanocrystal Quantum Dots. *Phys. Rev. B* **2001**, 63, 205316.
- [8] Tang, J.; Marcus, R. A. Mechanisms of Fluorescence Blinking in Semiconductor Nanocrystal Quantum Dots. *J. Chem. Phys.* **2005**, 123, 054704.

- [9] Verberk, R.; van Oijen, A. M.; Orrit, M. Simple Model for the Power-Law Blinking of Single Semiconductor Nanocrystals. *Phys. Rev. B* **2002**, 66, 233202.
- [10] Rosen, S.; Schwartz, O.; Oron, D. Transient Fluorescence of the Off State in Blinking CdSe/CdS/ZnS Semiconductor Nanocrystals Is Not Governed by Auger Recombination. *Phys. Rev. Lett.* **2010**, 104, 157404.
- [11] Pistol, M. E.; Castrillo, P.; Hessman, D.; Prieto, J. A.; Samuelson, L. Random Telegraph Noise in Photoluminescence from Individual Self-Assembled Quantum Dots. *Phys. Rev. B* **1999**, 59, 10725-10729.
- [12] Bruhn, B.; Valenta, J.; Sangghaleh, F.; Linnros, J. Blinking Statistics of Silicon Quantum Dots. *Nano Lett.* **2011**, 11, 5574-5580.
- [13] Cichos, F.; Martin J.; von Borczyskowski, C. Emission Intermittency in Silicon Nanocrystals. *Phys. Rev. B* **2004**, 70, 115314.
- [14] Bruhn, B.; Sangghaleh, F.; Linnros, J. Fabricating Single Silicon Quantum Rods for Repeatable Single Dot Photoluminescence Measurements. *Phys. Status Solidi A* **2011**, 208, 631-634.
- [15] Liu, H. I.; Biegelsen, D. K.; Johnson, N. M.; Ponce, F. A.; Pease, R. F. W. Self-Limiting Oxidation of Silicon Nanowires. *J. Vac. Sci. Technol. B* **1993**, 11, 2532-2537.
- [16] Kuno, M.; Fromm, D. P.; Hamann, H. F.; Gallagher, A.; Nesbitt, D. J. Nonexponential "Blinking" Kinetics of Single CdSe Quantum Dots: A Universal Power Law Behavior. *J. Chem. Phys.* **2000**, 112, 3117-3120.

- [17] Valenta, J.; Fucikova, A.; Vacha, F.; Adamec, F.; Humpolickova, J.; Hof, M.; Pelant, I.; Kusova, K.; Dohnalova, K.; Linnros, J. Light-Emission Performance of Silicon Nanocrystals Deduced from Single Quantum Dot Spectroscopy. *Adv. Funct. Mater.* **2008**, 18, 2666-2672.
- [18] Kuno, M.; Fromm, D. P.; Hamann, H. F.; Gallagher, A.; Nesbitt, D. J. "On"/"Off" Fluorescence Intermittency of Single Semiconductor Quantum Dots. *J. Chem. Phys.* **2001**, 115, 1028-1040.
- [19] Wang, S.; Querner, C.; Emmons, T.; Drndic, M.; Crouch, C. H. Fluorescence Blinking Statistics from CdSe Core and Core/Shell Nanorods. *J. Phys. Chem. B* **2006**, 110, 23221-23227.
- [20] Krauss, T. D.; O'Brien, S.; Brus, L. E. Charge and Photoionization Properties of Single Semiconductor Nanocrystals. *J. Phys. Chem. B* **2001**, 105, 1725-1733.
- [21] Murthy, A. V. R.; Patil, P.; Datta, S.; Patil, S. Photoinduced Dark Current due to Blinking and Photodarkening Probability of Aqueous CdTe Quantum Dots. *J. Phys. Chem. C* **2013**, 117, 13268-13275.
- [22] Early, K. T.; Nesbitt, D. J. Size-dependent Photoionization in Single CdSe/ZnS Nanocrystals. *Nano Lett.* **2013**, 13, 4844-4849.
- [23] Issac, A.; Krasselt, C.; Cichos, F.; von Borczyskowski, C. Influence of the Dielectric Environment on the Photoluminescence Intermittency of CdSe Quantum Dots. *Chem. Phys. Chem.* **2012**, 13, 3223-3230.
- [24] Hohng S.; Ha, T. Near-Complete Suppression of Quantum Dot Blinking in Ambient Conditions. *J. Am. Chem. Soc.* **2004**, 126, 1324-1325.

[25] Stefani, F. D.; Knoll, W.; Kreiter, M.; Zhong, X.; Han, M. Y. Quantification of Photoinduced and Spontaneous Quantum-dot Luminescence Blinking. *Phys. Rev. B* **2005**, 72, 125304.