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Microsecond Blinking Events in the Fluorescence of Colloidal Quantum Dots Revealed by Correlation Analysis on Preselected Photons

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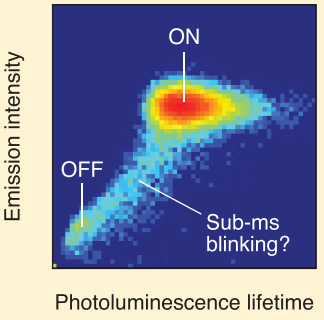
Supporting Information

ABSTRACT: Nearly all colloidal quantum dots, when measured at the single-emitter level, exhibit fluorescence "blinking". However, despite over 20 years of research on this phenomenon, its microscopic origins are still debated. One reason is a gap in available experimental information, specifically for dynamics at short (submillisecond) time scales. Here, we use photon-correlation analysis to investigate microsecond blinking events in individual quantum dots. While the strongly distributed kinetics of blinking normally makes such events difficult to study, we show that they can be analyzed by excluding photons emitted during long bright or dark periods. Moreover, we find that submillisecond blinking events are more common than one might expect from extrapolating the power-law blinking statistics observed on longer (millisecond) time scales. This result provides important experimental data for developing a microscopic understanding of blinking. More generally, our method offers a simple strategy for analyzing microsecond switching dynamics in the fluorescence of quantum emitters.



Tremendous progress has been made over the past 25 years[1] in improving the fluorescence of colloidal quantum dots (QDs) in terms of their brightness,[2] stability,[3,4] color purity,[5] and tunability via shape[6,7] and composition.[8,9] Nevertheless, blinking—random fluctuations in the fluorescence intensity between a bright "ON" level and dimmer "OFF" levels—remains a commonly observed phenomenon when individual quantum dots are studied. Despite intense investigation of this effect, the origin of blinking is still debated.[10,11] Random charging and discharging[12−14] of the QD by means of charge-carrier ejection from a doubly excited state (biexciton)[15] has been identified as a key component of the blinking mechanism. However, this process alone cannot explain why the kinetics of ON−OFF switching is non-exponential in nearly all samples.[11,16] The observation of distributed kinetics implies that ON−OFF switching is not due to a single process with a well-defined and constant rate. Consequently, various additional effects have been considered, including multiple charge-carrier trap states,[17,18] fluctuating tunneling barriers,[16] and spectral diffusion of intraband transitions.[19] With the right assumptions, all of these models can produce the experimentally observed distributed kinetics for ON−OFF switching. To what extent each of these models correctly describes the underlying microscopic mechanism of blinking remains unclear. It is possible that the blinking mechanism varies between different types of QDs or between different individual QDs from the same batch.[20−22] Multiple mechanisms could even be operative in the same individual QD.

Validation or refinement of existing models for QD blinking is challenging because the experimental characterization of the phenomenon is incomplete. Much work has focused on characterizing the *properties* of the ON and OFF states (and/or other intermediate-intensity states) for various types of QDs using time-correlated single-photon counting, including, e.g., the quantum efficiency, excited-state lifetime,[13,14] and multi-exciton emission properties.[23] In contrast, the *transitions* between the ON and OFF states are much more difficult to study. The statistics of these switching events are usually probed on millisecond time scales and longer by means of binning and thresholding[16,24] or change-point analysis.[25,26] However, the analysis of ON−OFF transitions is complicated by the finite excitation rates (typically lower than 1 μs^{-1}) and finite photon-collection efficiencies (typically lower than 10%) in experiments. Signal from an individual QD must therefore be accumulated for at least ∼1 ms before an ON−OFF switching event can be distinguished from a statistical fluctuation in the photon count rate. As a result, little is known about the statistics of ON−OFF transitions on submillisecond time scales.[16,24] Therefore, new strategies that can reveal microsecond blinking dynamics would be useful for testing and refining models for QD blinking. This is necessary to advance our microscopic understanding of this important phenomenon.

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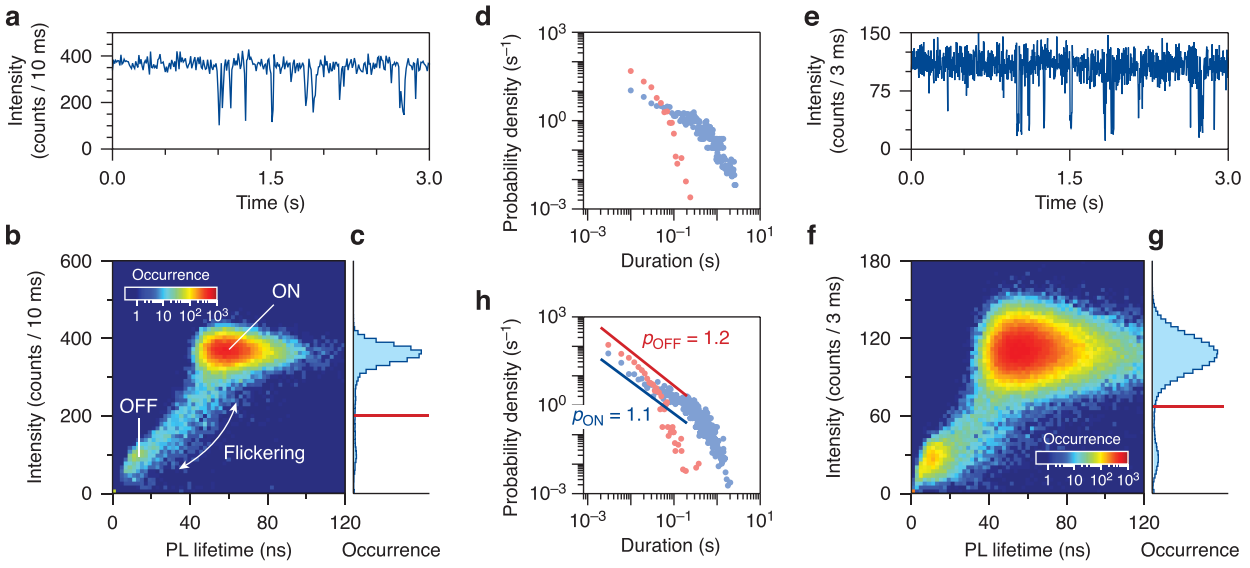

Figure 1. (a) Emission intensity from an individual CdSe/CdS/ZnS core/shell/shell QD binned with a time resolution of 10 ms under pulsed excitation (405 nm; 10 MHz; 1 μJ cm^{-2}). The QD core diameter is 3.2 nm,[39] and the shell has nominally 8 (2) monolayers of CdS (ZnS).[37] (b) Corresponding "fluorescence lifetime intensity distribution" (FLID), a two-dimensional histogram of the emission intensities and fitted fluorescence lifetimes.[40] The diagram is based on a 300 s experiment divided into 30 000 10 ms time bins (see Figure S1 in the Supporting Information for details). The effect of flickering is highlighted. (c) Corresponding one-dimensional intensity histogram, with the threshold used for statistical analysis indicated as a red line. (d) Blinking statistics for the ON periods (blue) and OFF periods (red), extracted from 10 ms binning and thresholding for the data in panel a.[16] (e–h) Same as panels a–d but obtained using 3 ms time bins on the same single-photon data.

Here, we demonstrate an analysis method to study the microsecond blinking dynamics of individual QDs. It is based on photon correlation, a technique that can in principle reveal subnanosecond dynamics (depending on the equipment used) and, consequently, is often used to study photon antibunching[27] or multiexciton emission[28−31] from single QDs. Typically, if a process exhibits widely distributed kinetics, as in the case of blinking, slow events (blinking on millisecond to second time scales) dominate the correlation function and fast events (blinking on microsecond time scales) are obscured.[32−35] To circumvent this issue, we first bin the single-photon data in 10 ms intervals and identify moments when the QD "flickers," i.e. when it switches between ON and OFF on a sub-10 ms time scale. By performing our correlation analysis only on preselected photons emitted during these moments, we exclude slow ON−OFF switching events. This allows us to reveal signatures of fast microsecond switching that are otherwise hidden. The experimental correlation functions are then compared with Monte Carlo simulations. We conclude that microsecond blinking events are more frequent than may be expected from an extrapolation of a power-law model for the millisecond blinking statistics. This seems to be inconsistent with the diffusion model for blinking,[35,36] while for other models it poses additional restrictions on the distributions and fluctuations of trapping and/or nonradiative recombination rates.

We begin by applying our method to high-quality CdSe/CdS/ZnS core/shell/shell QDs that were synthesized following the procedure of Boldt et al.,[37] which is related to a protocol from Chen et al.[38] It uses continuous injection of metal oleate (cadmium or zinc) and octanethiol at high temperature to obtain high-quality shells. Figure 1a shows 3 s of a typical emission intensity trace under pulsed excitation (405 nm; 10 MHz; 1 μJ cm^{-2}) of an individual QD with a 3.2 nm core diameter[39] and a shell nominally of 8 monolayers of CdS and 2 monolayers of ZnS.[37] Under continual excitation, the QD switches randomly between "ON" periods of high emission intensity and low-intensity "OFF" periods. However, it is ON most of the time. This is consistent with the "reduced blinking" behavior of these types of QDs reported in previous studies.[37,38]

The emission intensity trace of Figure 1a is binned at 10 ms resolution. For each of the 30 000 time bins in the 300 s experiment, we can construct a decay histogram for the 100−400 photons in the bin and determine the fluorescence lifetime using a maximum-likelihood fit routine (see Figure S1 in the Supporting Information for details). We can then analyze how, along with the emission intensity, the fluorescence lifetime fluctuates during the 300 s experiment. As demonstrated previously,[40] the results can be presented in a "fluorescence lifetime intensity distribution" (FLID) plot (Figure 1b), which is a two-dimensional histogram showing the correlations between emission intensity and fluorescence lifetime. We see that the QD is often in an "ON state" with a count rate of approximately 360 counts/10 ms and a fluorescence lifetime of ∼56 ns, and sometimes in an "OFF state" with approximately 100 counts/10 ms and a lifetime of ∼8 ns. The characteristics of these states are consistent with blinking due to random charging and discharging,[12,13,41,42] where the QD is neutral in the ON state and charged in the OFF state (Figure S2).

A common strategy for analyzing ON−OFF blinking dynamics is to assign each 10 ms time bin to an ON or OFF period based on an emission intensity threshold (red line in Figure 1c) and then histogram the durations of ON and OFF periods that occurred during the experiment (Figure 1d).[11,16] This analysis reveals the characteristic nonexponential dynamics of blinking, which often resemble a (truncated) power law.[11,15,18] However, it does not show blinking dynamics on time scales faster than the bin width of 10 ms. The FLID plot (Figure 1b) reveals that faster blinking events do occur during the experiment (during those moments in which the measured count rate and fluorescence lifetime are

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intermediate between that of the ON and the OFF states). These moments (highlighted in Figure 1b with a double-headed arrow) can be due to "flickering", i.e., rapid blinking resulting in one or more ON or OFF periods lasting shorter than 10 ms.[20,22]

An obvious way to improve the time resolution of the thresholding analysis is to use finer binning. In Figure 1e–h we repeat the above analysis on the same single-QD fluorescence data but with 3 ms binning. This enables us to resolve some fast blinking events that were initially hidden, for example the short OFF period at t = 0.3 s (compare Figure 1a to Figure 1e). However, this comes at the cost of a diminished signal-to-noise ratio, as is clear from the broader peaks in the distributions extracted from the count rates and fluorescence lifetimes (compare Figure 1b,c to Figure 1f,g). More importantly, while the thresholding analysis of Figure 1h confirms that blinking events on time scales between 3 and 10 ms are frequent, the FLID plot (Figure 1f) still shows effects of flickering due to unresolved ON–OFF transitions. This indicates that 3 ms binning is still insufficient to characterize the fastest blinking events. Indeed, extrapolating the blinking statistics of Figure 1h and assuming approximate power-law behavior would even suggest that fast blinking events (microsecond to millisecond) are more frequent than slower events (>ms). Because of the increasing noise at finer binning (compare Figure 1a–c to Figure 1e–g), these fast events are difficult to quantify using conventional threshold analysis or more advanced methods (such as change-point analysis) that rely on directly identifying ON–OFF transitions from variations in the photon count rate (see also Figure S3). Also, power-spectral density analysis of blinking is limited to low frequencies (~kHz and slower) because of Poissonian noise.[35,43]

The intensity correlation function

$$g^{(2)}(t) = \frac{\langle I(t_0)I(t_0 + t)\rangle}{\langle I(t_0)\rangle\langle I(t_0 + t)\rangle} \quad (1)$$

has been proposed as a route to analyze the fast dynamics of blinking.[32−35] Here, $I(t_0)$ is the emission intensity at time t_0 and $I(t_0 + t)$ is the intensity some time t later; $\langle\bullet\rangle$ denotes averaging t_0 over the entire experiment. Indeed, the correlation function is routinely used to reveal photon antibunching on nanosecond time scales, which can be done with a high signal-to-noise ratio provided the experiment is sufficiently long. The intensity correlation function (Figure 2a) for our measurement on the nanosecond time scale shows a series of peaks at regular time intervals, as expected under pulsed excitation.[27] The peaks are strongly overlapping here because of our fast repetition rate (10 MHz) compared to the excited-state lifetime of the QD under study (60 ns in the ON state). Nevertheless, clear antibunching, i.e., a dip in the correlation function at zero time delay, proves that we are studying a single QD. We chose pulsed excitation with a relatively fast repetition rate to optimize the count rate while simultaneously allowing us to track lifetime fluctuations in the QDs. The correlation analysis we present in this work does however not require pulsed excitation and works equally well with continuous-wave excitation. Figure 2b shows the intensity correlation function over a wider range of time scales from 10 ns to 1 s. At t > 1 ms, the function approaches a value of 1. It has a slightly higher value of approximately $g^{(2)} \approx 1.055$ over 3 orders of magnitude in time between 1 μs and 1 ms, as has been previously

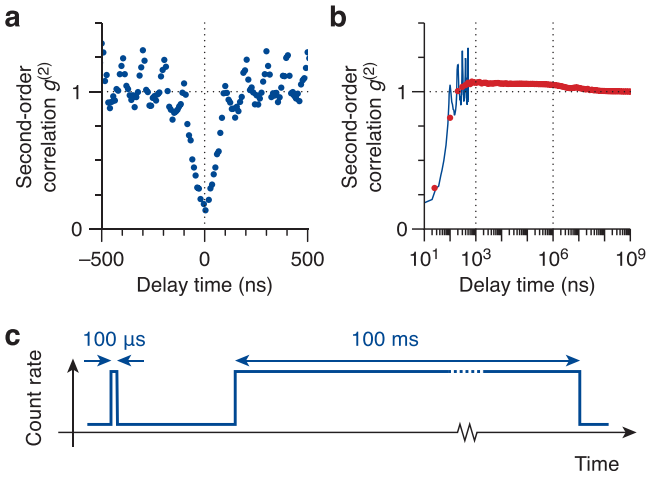

Figure 2. (a) Intensity correlation function $g^{(2)}$ constructed from the same experiment as analyzed in Figure 1. Antibunching with $g^{(2)}(t = 0) < 0.5$ proves that we are studying a single emitter. (b) The same intensity correlation function rebinned to integer numbers of 100 ns laser repetition periods (red data points), plotted with a logarithmic time axis. Photon pairs with positive (detector 1 clicks before detector 2) and negative (vice versa) delay times have been averaged, and both are plotted at positive delay times. The blue line shows the first 500 ns at high resolution (same as in panel a). (c) Schematic (without Poisson noise) of the situation of a typical blinking experiment, in which the duration of ON periods varies widely. The correlation function is basically a histogram of time differences between photon pairs, so it is dominated by the long periods. In this example, the 100 ms ON period contributes more photon pairs than the 100 μs ON period by a factor of 10^6.

observed.[32] To interpret the intensity correlation function, we note that the value of $g^{(2)}(t)$ reflects how "similar" the QD is on average at time $t_0 + t$ compared to at time t_0 when it emitted a photon. In other words, our observation that $g^{(2)}$ slowly decreases toward unity for t > 1 ms reflects changes of the QD state—i.e., blinking—on time scales of milliseconds and longer.

On shorter time scales, the plot in Figure 2b contains very limited information about blinking, as is clear from the nearly flat and featureless shape of the curve for 1 μs < t < 1 ms. This does not necessarily mean that submillisecond blinking does not occur, but can instead be a consequence of the peculiar nonexponential dynamics of blinking (Figure 1d,h), as discussed previously.[35] These cause short blinking events to be hidden by the contributions from slower and much brighter events (Figure 2c). Clearly, an alternative strategy is necessary to extract information about submillisecond blinking dynamics.

The method we propose and apply here to reveal submillisecond blinking dynamics is conceptually simple. Instead of constructing the correlation function $g^{(2)}$ for the entire experiment (in this case, 10^7 photons collected over 300 s), we focus on moments during which the QD shows an intermediate emission intensity in the binned data. For example, we select all 10 ms time bins during which 124–160 counts are recorded (yellow regions in Figure 3a,b), compared to averages of 100 counts/10 ms for the OFF state and 360 counts/10 ms for the ON state. For these moments, the QD may have exhibited rapid blinking on a sub-10 ms time scale, as discussed above (Figure 1b,f). A total of 65 000 photons were recorded during these moments. We then calculate the intensity correlation function of these preselected

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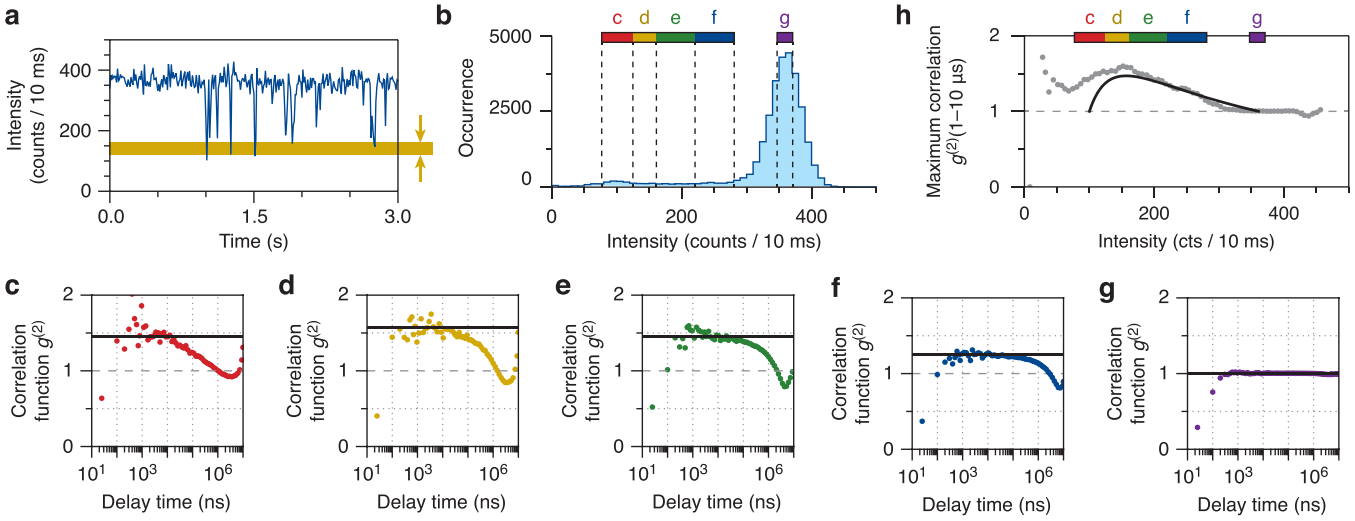

Figure 3. (a) Emission intensity trace of the QD (reproduced from Figure 1a) with a range of intensities highlighted in yellow (120–160 counts/10 ms) that are likely due to flickering, i.e. ON–OFF switching on time scales faster than the 10 ms binning used. (b) Intensity histogram with different intensity ranges highlighted that are used to preselect photons for correlation analysis. (c–g) Correlation functions $g^{(2)}$ constructed from preselected photons emitted during 10 ms time bins in which 76–124 (red; c), 124–160 (yellow; d), 160–220 (green; e), 220–280 (blue; f), or 348–372 (purple; g) photons were recorded. (h) Maximum bunching amplitude, i.e., $g^{(2)}$(1–20 μs), as a function of the condition for emission intensity that was used to preselect photons for the analysis.

photons (Figure 3d). It shows a negative slope on the microsecond time scale, from $g^{(2)}(1\ \mu s) = 1.5$ to $g^{(2)}(1\ ms) = 1.1$, a signature of blinking on these microsecond–millisecond time scales. Hence, while fast blinking dynamics are hidden in the intensity correlation function of the total experiment (Figure 2b), they are revealed when we construct the correlation function on preselected photons emitted during flickering periods. In addition, we observe antibunching at t = 2–6 ms (i.e., $g^{(2)} < 1$ in Figure 3d) and strong bunching at t = 10 ms (i.e., $g^{(2)} > 1$ in Figure 3d). These features do not reflect surprising blinking dynamics of the QD but instead are "finite-bin-size" artifacts arising because we analyze time bins of 10 ms (see Figures S4 and S5).

We can systematically vary the condition by which we select photons for our correlation analysis. We plot five intensity correlation functions, constructed from 10 ms time bins with photon counts of 76–124 (red; Figure 3c), 124–160 (yellow; Figure 3d), 160–220 (green; Figure 3e), 220–280 (blue; Figure 3f), or 348–372 (purple; Figure 3g). The intensity ranges considered are also highlighted with different colors in the intensity histogram of Figure 3b. The correlation function during ON periods (Figure 3g) is completely flat except for the antibunching feature at nanosecond time scales. Hence, when the QD yields 348–372 photon counts in a 10 ms time bin, it is in a stable well-defined ON state, and no sub-10 ms blinking events occur. The other plots (Figure 3c–f) show bunching (i.e., $g^{(2)} > 1$), indicating intensity fluctuations (i.e., blinking) on the sub-10 ms time scale. $g^{(2)}$ peaks at $t_{max} \approx 1$–20 μs, as highlighted with black horizontal lines that show the average of $g^{(2)}$ over this range of delay times. Note that the intensity histogram constructed with 3 ms binning (Figure 1g) does not present a clean range of intermediate intensities. As a result, the above selection procedure of flickering events could not be performed based on the histogram binned at 3 ms.

Assuming that the bunching is due to two-state blinking between an ON and an OFF state with intensities I_{ON} and I_{OFF}, we would expect the maximum bunching amplitude for time scales t_{max} that are faster than the fastest blinking events (but slower than the excited-state lifetime) to satisfy

$$g^{(2)}(t_{max}) = \frac{\langle I^2 \rangle}{\langle I \rangle^2} = \frac{I_{ON} + I_{OFF}}{\langle I \rangle} - \frac{I_{ON} I_{OFF}}{\langle I \rangle^2} \quad (2)$$

where $\langle \bullet \rangle$ denotes averaging over the time bins selected to construct the correlation function (see the Supporting Information for a derivation). Figure 3h shows that the experimental bunching amplitude (gray data points) follows eq 2 with constant I_{ON} and I_{OFF} (black line), except at low emission intensity. This means that blinking of this QD is only approximately binary, i.e. between two states. The experimental results are consistent with a situation in which the QD blinks between an ON state with I_{ON} = 360 counts/10 ms and multiple low-intensity OFF states with similar emission intensities of $I_{OFF} \approx$ 100 counts/10 ms. This scenario explains that even if we preselect time bins of ~100 counts/10 ms (i.e., moments when the QD is in its OFF state), the correlation function is not flat but instead shows clear effects of fluctuations on submillisecond time scales (Figure 3c). This indicates that what appears to be an OFF state in the FLID plots of Figure 1b,f is in fact not a single well-defined state in which the QD is stable for several milliseconds. Instead, submillisecond intensity fluctuations occur when the QD emits at approximately 100 counts/10 ms.

Now that we understand the bunching amplitude in the correlation function of preselected photons, we turn to the dependence of bunching on delay time. This provides information about the statistics of fast blinking events in the QD. The simplest flickering scenario for a 10 ms time bin in which the QD displays some intermediate intensity (e.g., 124–160 counts) is that it was mostly off (I_{OFF} = 100 counts/10 ms) but exhibited a single short ON period (I_{ON} = 360 counts/10 ms) of 1.5 ms. This scenario, highlighted in yellow in Figure 4a, would yield an expectation value of $\langle I \rangle$ = 139 counts. Figure 4b shows the expected intensity correlation function for this scenario (black line; see Figure S4 for details of the calculation) and compares it with experiment. Qualitatively,

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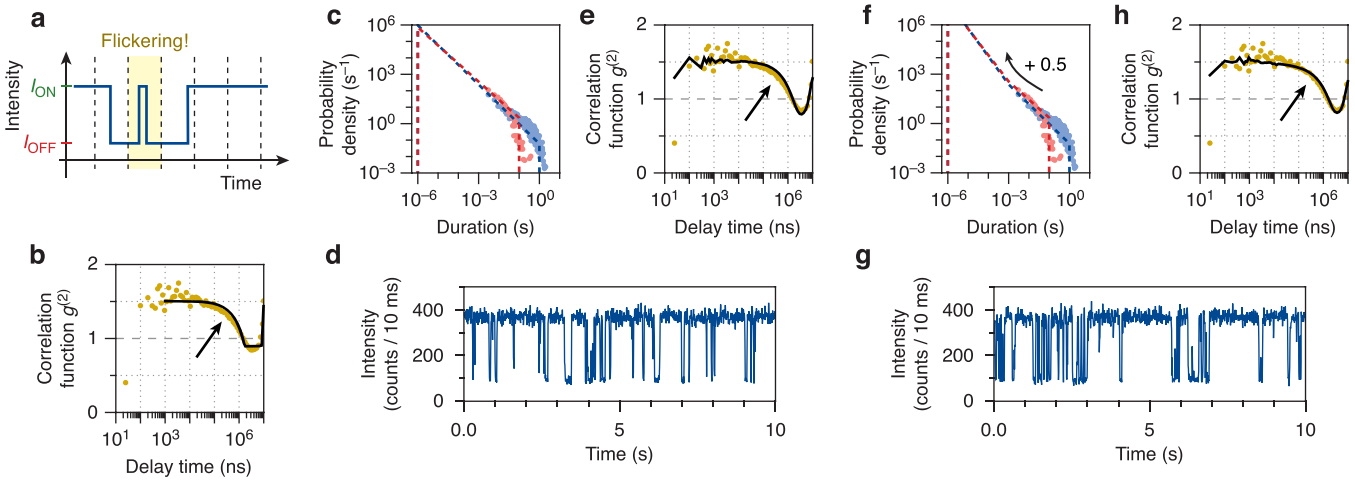

Figure 4. (a) Schematic of an emission intensity trace from an individual QD without Poisson noise. Vertical dashed lines indicate the time binning used. A flickering event is highlighted in yellow, i.e. blinking to an ON period shorter than the experimental binning. (b) Calculated correlation function $g^{(2)}$ for time bins that exhibit flickering such as that schematically shown in panel a (black line) compared to the experiment (data points, reproduced from Figure 3d). The correlation function is averaged over the timing of the short ON period with respect to the beginning of the time bin (details in Figure S4). The arrow highlights a deviation in the slope of the correlation function between the calculation and the experiment. (c) Blinking statistics assumed in our Monte Carlo model for single-QD emission for ON (blue) and OFF (red dashed line) periods, extrapolating the power-law experimental statistics obtained from binning and thresholding (data points) and cutting off at $t = 1$ μs at the short-time-scale end and at $t = 100$ ms (OFF) or $t = 1$ s (ON) at the long-time-scale end. See the Supporting Information for the Monte Carlo modeling procedure and more simulated correlation functions (Figure S5). (d) Part of a simulated intensity trace. (e) Correlation function of preselected photons from the simulation (black line), treated in the same way as the experimental data (points). The arrow highlights a deviation in the slope of the correlation function between the calculation and the experiment. (f–h) Same as panels c–e, but assuming that the ON and OFF statistics are described by a power-law dependence with steeper slope for 1 μs < t < 1 ms than for t > 1 ms. More precisely, the power-law exponents are $p_{ON1} = 1.7$ and $p_{OFF1} = 1.6$ for 1 μs < t < 1 ms and $p_{ON2} = 1.2$ and $p_{OFF2} = 1.1$ for t > 1 ms.

the calculation reproduces the artifacts due to the finite bin size, but the slope of the experimental correlation function at submillisecond time scales is steeper (highlighted by a black arrow). This suggests that the average intensity of $\langle I \rangle = 124$–160 counts in the experiment is due to several fast submillisecond flickering events, instead of the single ON period of ∼1.5 ms assumed for this calculation (Figure 4a).

To obtain a better match between the calculated and experimental correlation function, we simulated QD emission with a Monte Carlo model. We can extrapolate the experimental blinking statistics on the 3 ms time scale (Figure 1h) to shorter time scales assuming that the approximate power-law dependence (probability density $\propto t^{-p}$) is valid down to a microsecond (Figure 4c) and use this as input for our model (see Figure S6). This yields an intensity trace (Figure 4d) similar to the experiment. We then perform the same analysis that we did above for the experimental data on the simulated data. However, the corresponding correlation function from moments of QD flickering (Figure 4e; black line) still does not match the slope observed in the experiment (highlighted by a black arrow). We can improve the match further by assuming that the blinking on submillisecond time scales is *more* frequent than expected from simply extrapolating the power-law statistics observed at millisecond–second time scales. Panels f–h of Figure 4 show the simulation results obtained assuming that the blinking statistics can be described with two different power-law dependencies on different time ranges, where the power-law exponent p for both the ON and OFF duration is 0.5 higher (e.g., $p = 1.7$ versus 1.2) for the range $t = 1$ μs to 1 ms compared to the range $t = 1$ ms to 1 s. This model produces a slope in the intensity correlation function during flickering that is similar to the experiment (Figures 4h and S5). Although the differences between the corresponding correlation functions are subtle (compare panels b, e, and h of Figure 4), our analysis indicates that intensity fluctuations on submillisecond time scales are more common than one might expect from extrapolating the power-law statistics extracted from binning and thresholding (compare panels c and f of Figure 4). Our model with a change in power-law slope is inspired by Pelton et al.,[35] who provided evidence for a diffusion-controlled mechanism of blinking[36] in the form of a *reduced* power-law exponent for the blinking statistics on submillisecond time scales. Our analysis however shows an *increased* power-law slope on the shorter time scales. This apparent contradiction may in part arise because we use different (CdSe/CdS/ZnS versus CdSe/ZnS in ref 35) with significantly improved blinking behavior after several years of synthesis optimization.[3−5,37] Indeed, our QDs do not exhibit the same kink in the power spectrum of the intensity trace that Pelton et al. previously observed[35] (Figure S7).

Qualitative inspection of the correlation function of preselected photons (in particular Figure 3d,e) and the more quantitative analysis of Figure 4 demonstrate significant submillisecond blinking in the individual QD under study. We obtained similar results on individual QDs from three separate synthesis batches of CdSe/CdS/ZnS QDs,[37] as well as on two batches of CdSe/CdS dot-in-rods[44] (Figure S8). Unfortunately, correlation analysis cannot distinguish between short ON or OFF periods, so we cannot precisely deduce the ON and OFF statistics nor verify exactly how well the model of two different power-law slopes captures them. Nevertheless, our results give an indication that submillisecond intensity fluctuations, hidden by Poisson noise in traditional binning and thresholding analysis (Figure 1), are frequent in many QDs. On the other hand, in the same batches of CdSe/CdS/ZnS

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QDs and CdSe/CdS dot-in-rods we also found examples of emitters with quite different blinking behavior. These emitters exhibited relatively stable intermediate-intensity states (Figure S9) and associated intermediate photoluminescence lifetimes (Figure S10). The presence of these states could be identified with our correlation method, even if they were not immediately evident from standard binning analysis or from the FLID plot. Our findings are consistent with previous studies that found qualitatively different blinking behavior between nominally identical QDs from the same batch.[20−22] Therefore, detailed analysis methods like the one we introduce here are indispensable for distinguishing between different emission properties of individual QDs from the same batch.

To summarize, we have proposed and demonstrated a method—correlation analysis on preselected photons—for extracting information about the microsecond blinking dynamics of individual QDs. While the intensity correlation function of a full experiment is nearly flat and featureless on time scales from microseconds to milliseconds, rejecting photons emitted during long bright periods from the analysis reveals bunching due to fast flickering. Our results indicate that intensity fluctuations on the microsecond time scale are more frequent than may be expected from extrapolating the blinking statistics extracted from conventional binning and thresholding. The method presented here can fill a gap in the available data on QD blinking, between the nanosecond time scales of photoluminescence decay measurements[13,45] and the millisecond time scales of binning and thresholding.[16,24] This is important not only for the microscopic understanding and suppression of blinking in conventional CdSe-based QDs but also for newer materials based on InP, $CuInS_2$, or lead halide perovskites.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.jpclett.9b01348.

Description of the optical experiments and the Monte Carlo simulations, additional experimental data on individual quantum dots and dot-in-rods, and additional simulation results (PDF)

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Notes

The authors declare no competing financial interest.

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