Unbiased determination of single quantum dot blinking power-law exponents using the autocorrelation function

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Blinking, that is to say intermittent fluorescence, is a ubiquitous feature of the emission of nanoparticles and can have dramatic consequences for many potential applications, such as lasers, light emitting diodes, and single photon sources. Photoluminescence intermittence manifests itself as intensity fluctuations in the fluorescence timetrace of nano-emitters, where highly emissive states (ON states) are repeatedly interrupted by poorly emissive states (OFF states). The durations of these alternating ON and OFF periods are found to be distributed according to power laws for many kinds of quantum emitters, including CdSe/CdS quantum dots. Efforts to fully understand such blinking dynamics have been hampered by the difficulty of determining the exact power-law exponents from experimental intensity timetraces in the presence of noise; in fact, it has been shown by Crouch et al. that significantly different power-law exponents are obtained from one and the same stream of photons, depending on how the timetrace is binned before a subsequent threshold-based analysis tries to identify the ON and OFF periods [1].

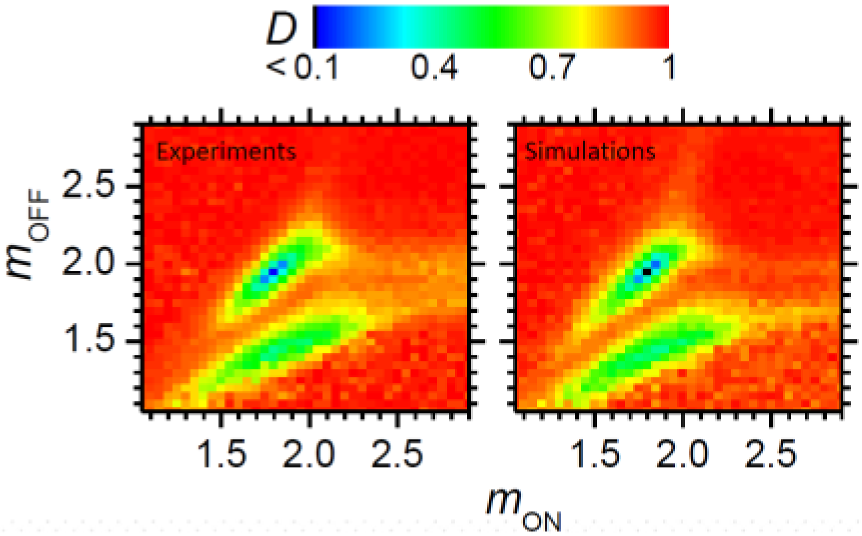

Fig. 1 Results of the Kolmogorov-Smirnov test: statistical parameter *D* as a colorscale, as a function of the ON and OFF power-law exponents. Smallest value of *D* indicates best fit to the experimental data. Left: experimental results. Right: simulated results.

We present an unbiased and robust analysis method for power-law blinking statistics in the photoluminescence of single nano-emitters, allowing us to extract both the bright- and dark-state power-law exponents from the emitters' intensity autocorrelation functions [2]. As opposed to the widely-used threshold method, our technique therefore does not require discriminating the emission levels of bright and dark states in the experimental intensity timetraces. We rely on the simultaneous recording of 450 emission timetraces of single CdSe/CdS core/shell quantum dots at a frame rate of 250 Hz with single photon sensitivity. Under such experimental conditions, our approach can determine ON and OFF power-law exponents with a precision of 3% from a comparison to numerical simulations, even for shot-noise-dominated emission signals with an average intensity below 1 photon per frame and per quantum dot.

In order to probe the blinking statistics at higher frequencies (close to the exciton lifetime ideally) and to get rid of potential sample inhomogeneity, we have transposed our method to the study of a single nano-object with sub-microsecond resolution. From the autocorrelation function of a single CdSe/CdS quantum dot recorded for 10 minutes, we are able to infer the power-law blinking exponents over 5 orders of magnitude, from 1 MHz to 10 Hz frequency range.

To the best of our knowledge, this is the only method able to determine, in an unbiased way, the blinking power-law exponents of the ON and OFF states independently, at the microsecond timescale.

1) C.H. Crouch *et al., Nano Lett.,* **2010,** *10*, 1692.
2) J. Houel *et al., ACS Nano,* **2015**, *9*, 886.