

The Influence of Continuous vs. Pulsed Laser Excitation on Single Quantum Dot Photophysics

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SUPPORTING INFORMATION

Preparation of QD films

Invitrogen organic capped QDs with 585 nm emission maximum (Qdot585 ITK Organic Quantum Dot) were washed once according to a modified version of Invitrogen's recommended washing procedure¹. In addition to the 75/25 solution of methanol and isopropanol, two drops of butanol were added to achieve precipitation in the initial step. After centrifugation, the QD pellet was resuspended in semiconductor grade toluene (Alpha Aesar). The QD suspension was diluted to 20pM with a solution of 0.1% polystyrene (Sigma-Aldrich, molecular weight 35,000) in semiconductor grade toluene. A 5 μ L drop of the dilute QD suspension was spun onto a quartz microscope coverslip at 3000 rpm for 60 s. Films were prepared the day of the experiment.

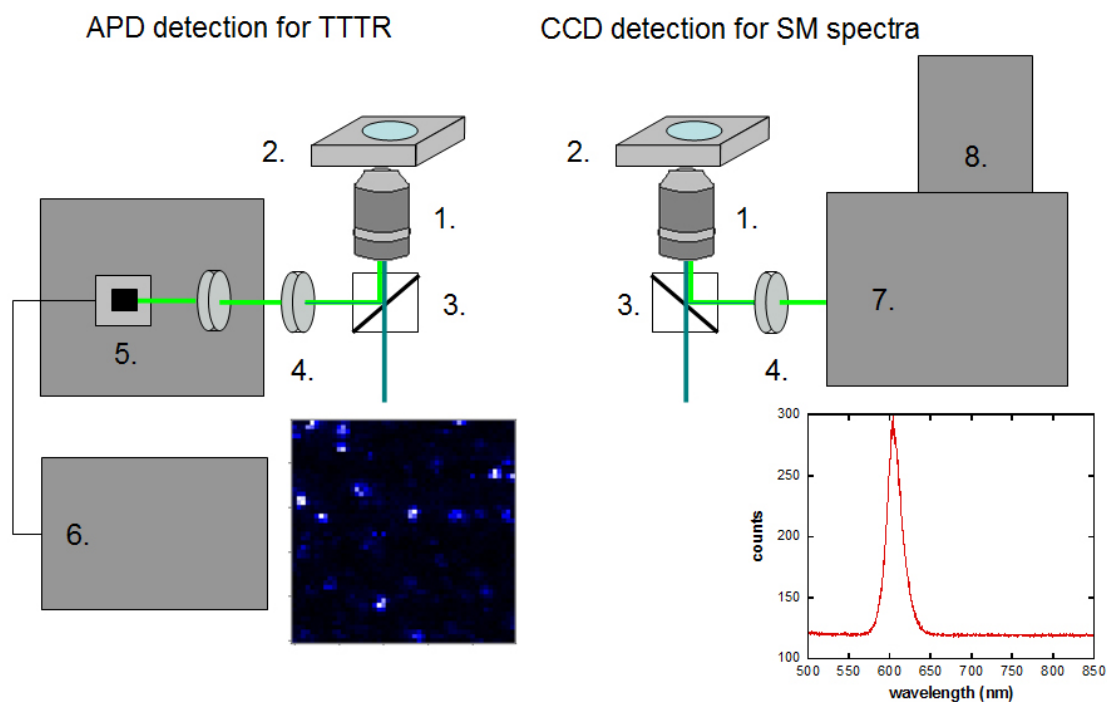


Fig. SI.1 Confocal Microscope setup (Nikon TU-3000)

1. Nikon Plan Fluor 100x oil immersion objective
2. Piezoelectric scanning stage
3. Chroma dichroic beamsplitter
4. Semrock Razor Edge filter (x2 for TTTR)
5. APD
6. PicoHarp
7. Acton Spectrometer
8. Princeton Instruments Spec-10 CCD

APD output (left), confocal image of QD intensities
 Spec-10 output (right), single-molecule QD spectrum

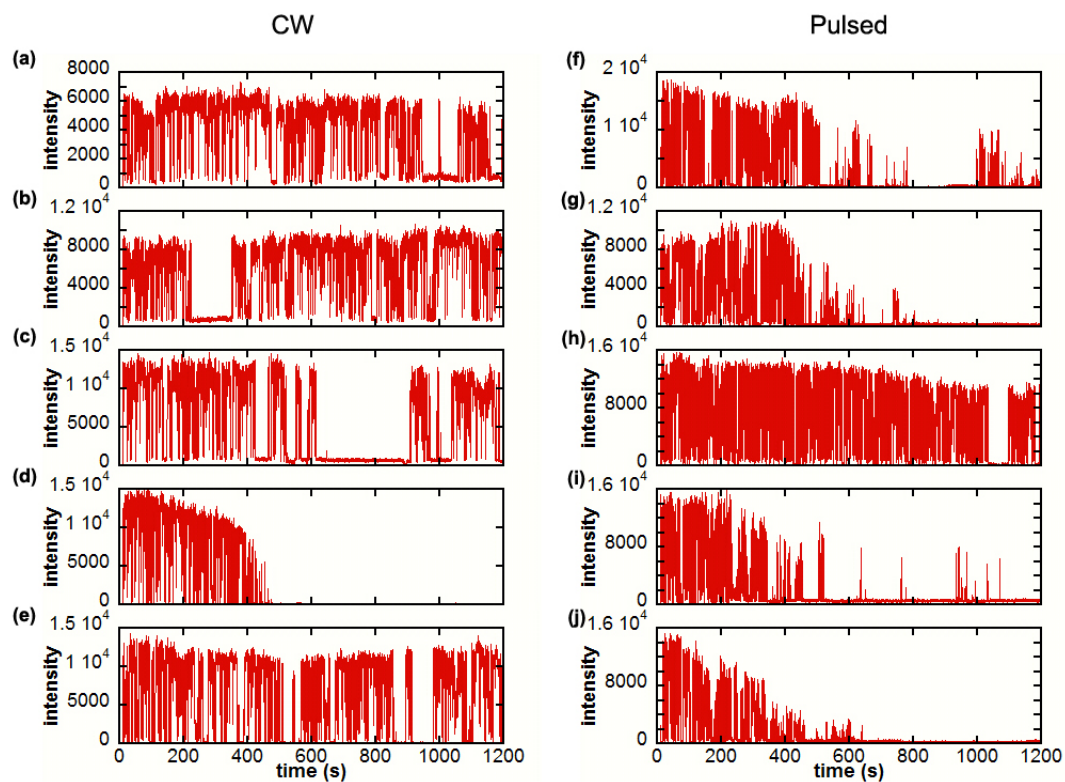


Figure SI.2 Representative single QD intensity traces are shown, for CW (a-e) and pulsed (f-j) excitation, with 100 ms bins.

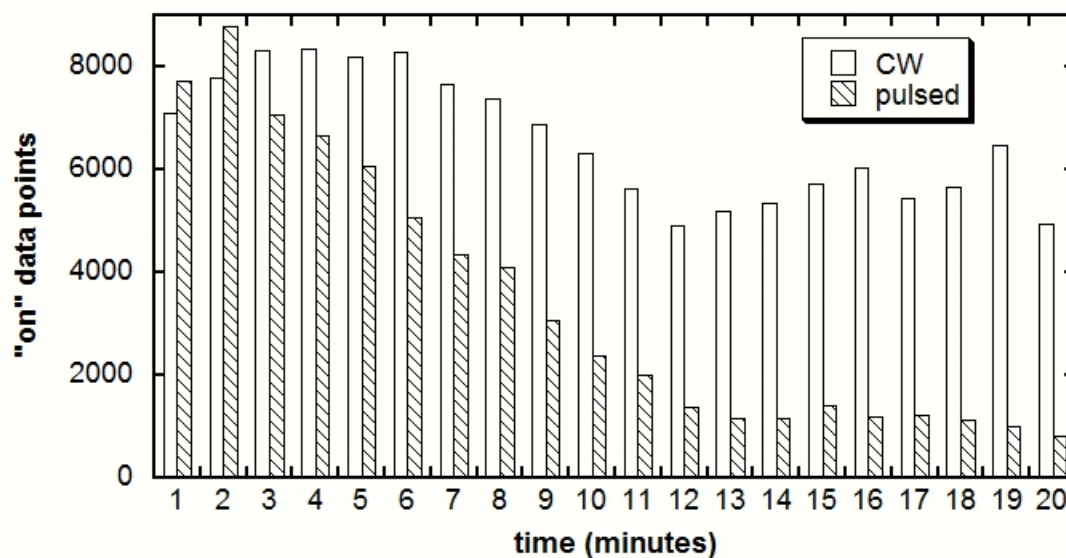


Figure SI.3 Blinking intensity traces from 40 QDs (20 with CW excitation and 20 with pulsed excitation) were selected at random, and data points above the on/off threshold were counted for each minute of the 20 minute experiment window. The pulsed data set (shaded bars) shows a drop off in number of ON events as compared to the CW data set (white bars) within the first few minutes of the experiment.

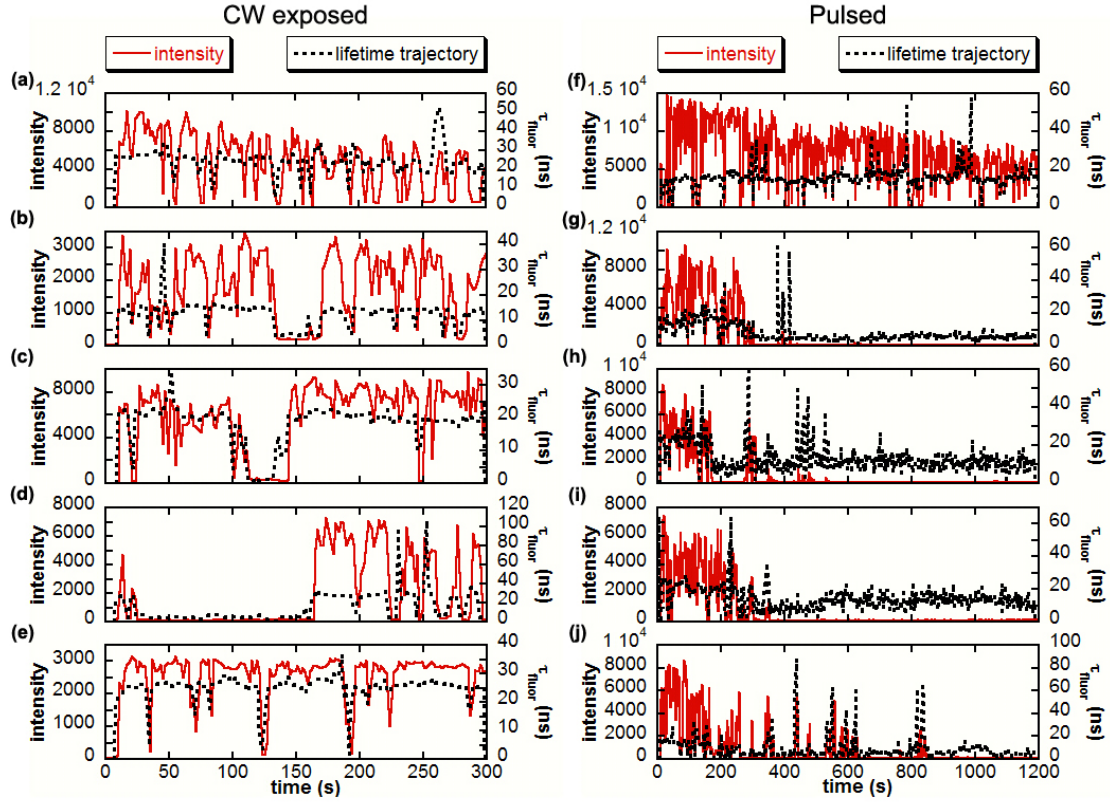


Figure SI.4 QD fluorescence intensity trajectories (solid line, left y-axis) are overlaid with the calculated lifetime trajectories (dashed line, right y-axis). CW exposed (a-e) and pulsed (f-j) traces are shown. Note that CW exposed data was only acquired for a 5 minute experiment window, compared to the full 20 minutes for the pulsed data set, and both are displayed with 2s bin time.

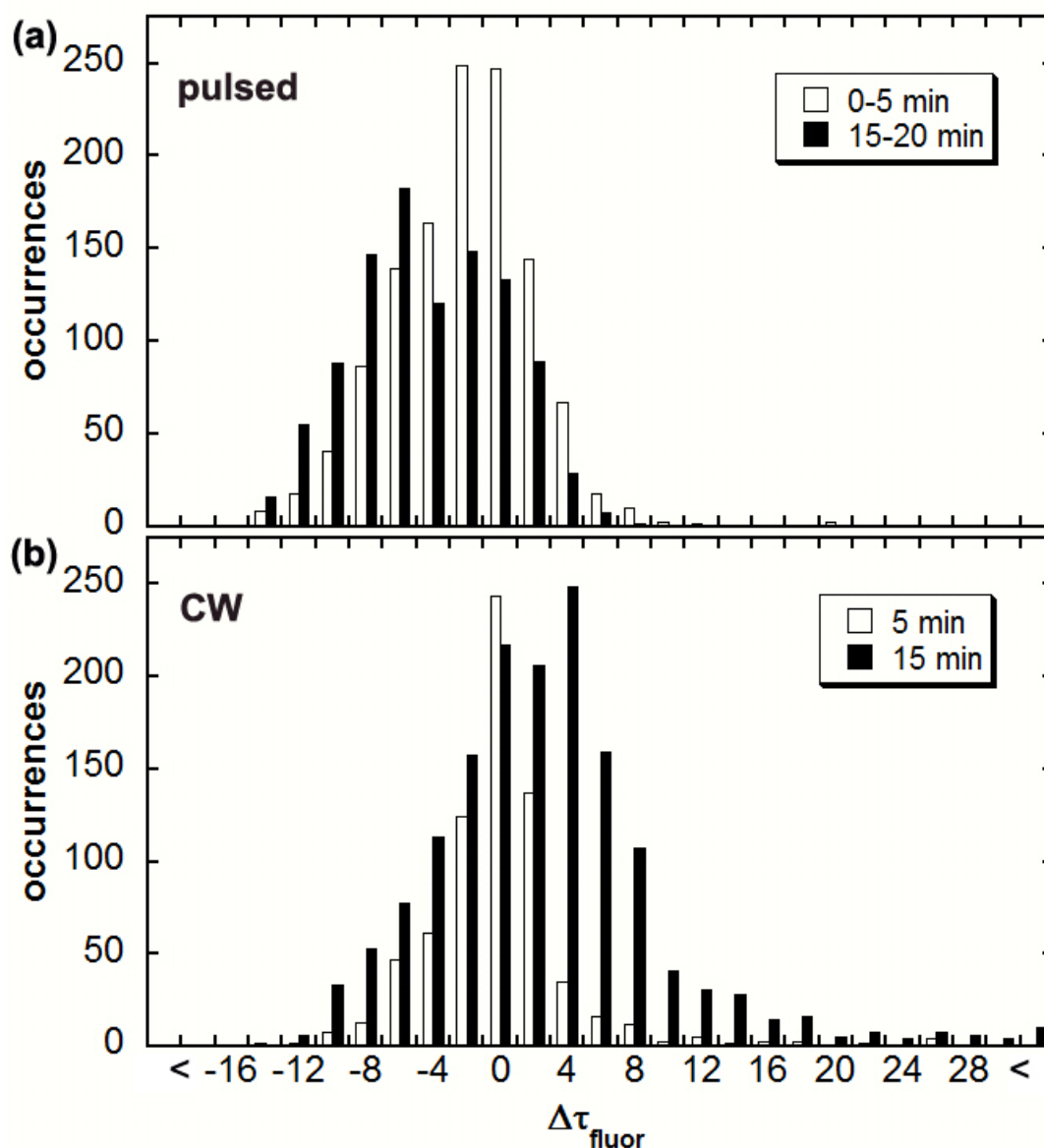


Figure SI.5 Lifetime deviations ($\Delta\tau_{fluor}$) are presented as histograms, representing a randomly selected portion of the pulsed data (10 QDs) and the CW exposed data. (a) For the pulsed data set, comparing the first and last 5 minute intervals of the full 20 minute experiment window shows a shift to shorter fluorescence lifetimes (negative $\Delta\tau_{fluor}$ values) with longer durations of excitation. (b) CW exposed data set is presented for 5 and 15 minutes of CW exposure, followed by 5 minutes of pulsed excitation for fluorescence lifetime measurements. Longer durations of CW excitation do not appear to result in a shift to shorter fluorescence lifetimes.

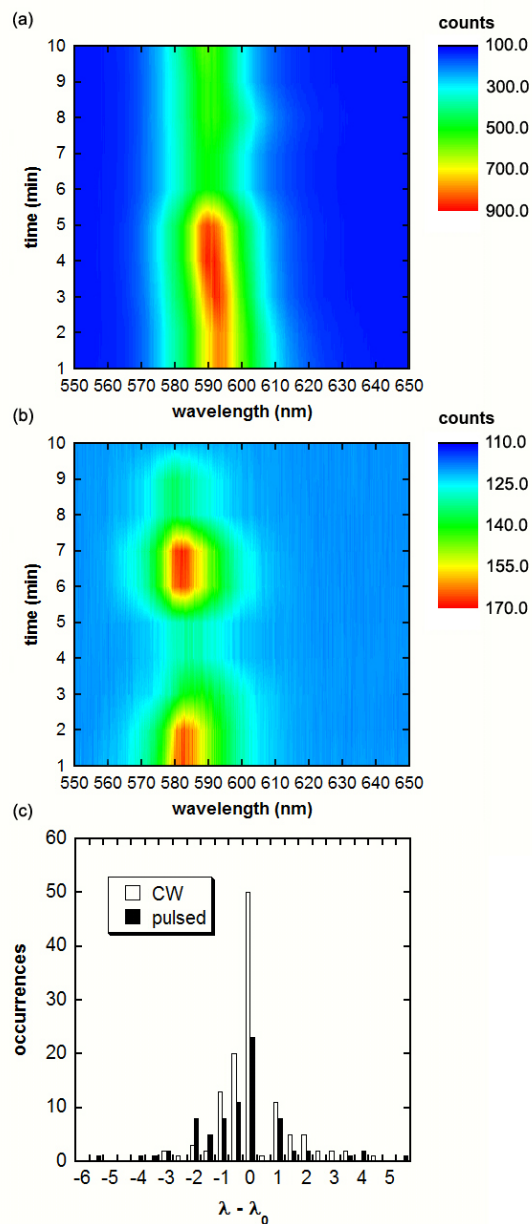


Figure SI-6. Single molecule fluorescence spectra were collected with a CCD camera to determine if the different types of excitation would influence spectral diffusion. Spectra were collected with 1 minute integration times, sequentially for 10 minutes for each QD. 12 QDs were excited by CW, and a representative spectral map is shown in (a), and 10 excited by pulsed, as represented in (b). The range of peak shift values ($\lambda - \lambda_0$) shown in (c) is very slightly larger for pulsed excitation (black bars) than CW excitation (white bars). Neither data set shows a tendency to either red or blue shift.

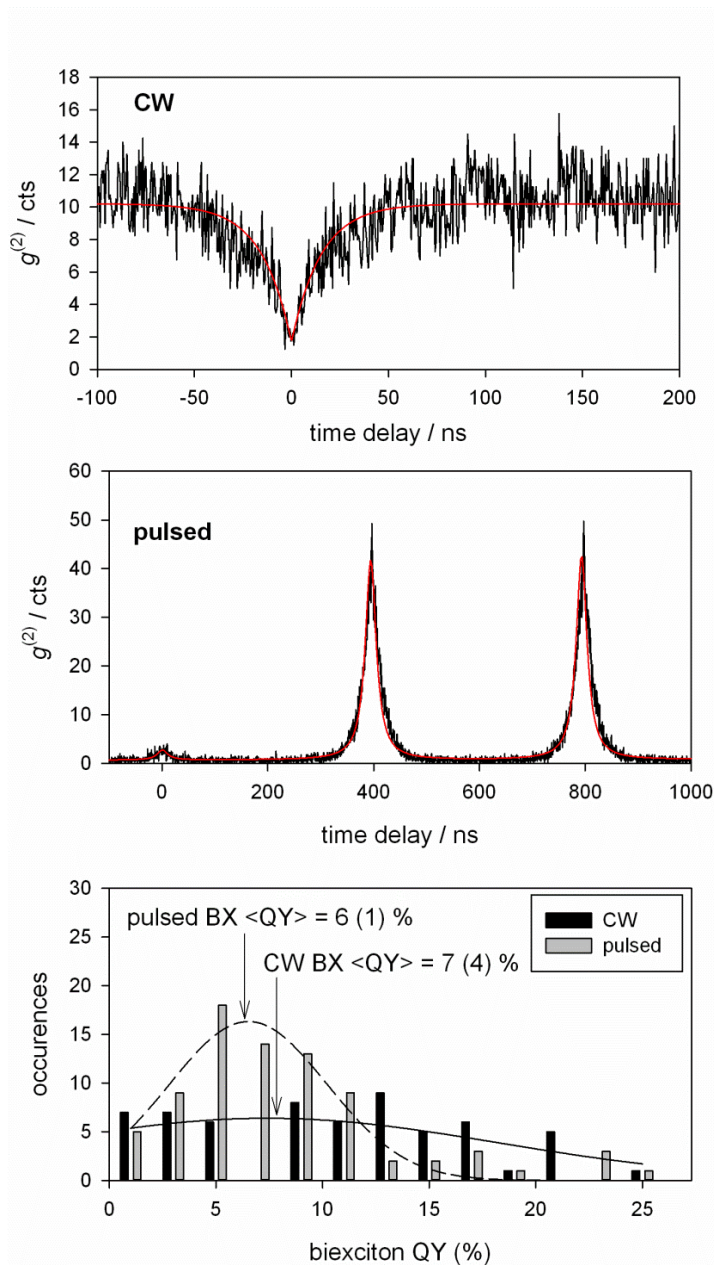


Figure SI-7. Example $g^{(2)}$ data for cw (top panel) and pulsed (middle panel) excitation that are used to calculate the biexciton fluorescence quantum yield (BX QY). Histograms (bottom panel) of the BX QY calculated for cw (black bar) and pulsed (grey bar) excitation.

Reference

1. "Flocculation of Qdot ITK Organic Quantum Dots from Decane." *Qdot Probe Manuals and Application Notes*. Life Technologies, 5 Dec. 2005. Web. 27 Jan. 2014. <<http://tools.lifetechnologies.com/content/sfs/manuals/mp19023.pdf>>.