Supporting Materials

Luminescence Dynamics of Silica Encapsulated Quantum Dots During Optical Trapping

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Inhomogeneous emission broadening of trapped QD clusters

To confirm the trapping of QD ensembles of bunched nanoparticles (or clusters) under our experimental conditions, inhomogeneous broadening of trapped nanoparticles emission has been studied. As reported in Ref. S1, the room temperature emission of a typical single QD presents a narrow peak of about 10 nm width. On the other hand, the spectral position of the peak dramatically depends on the confinement energy of the photo-exciton, that is, on the size of the nanocrystal. Since bulk QD@SiO_2 and pQD@SiO_2 samples present a normal size distribution, the emission peak broadens to a Gaussian profile. Regardless of the sample concentration and laser power, the emission width collected from our optical trap is always about 40 nm, and it equals the inhomogeneous broadening measured from bulk QD@SiO_2 and pQD@SiO_2 samples in a fluorometer (see Fig. S1 for an example).

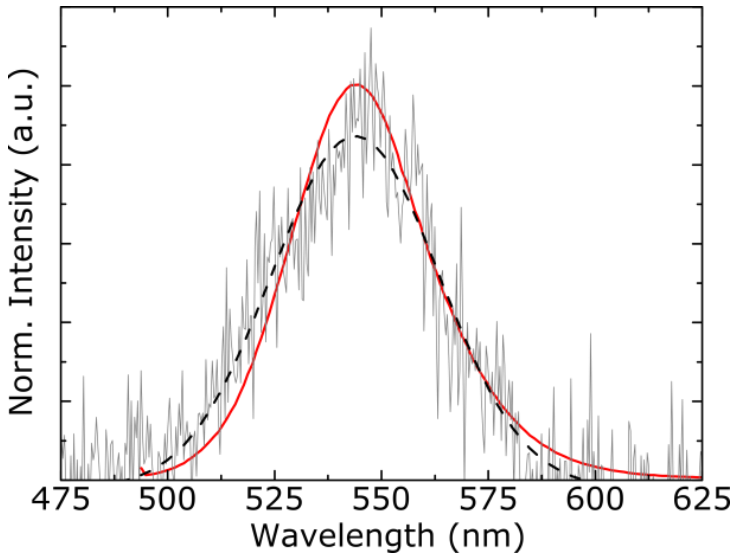

Figure S1. Emission spectra from bulk pQD@SiO_2 (red), from an optically trapped pQD@SiO_2 cluster (grey) and Gaussian fitting to cluster emission (dashed curve) showing a peak width of 39.6±0.9 nm.

Multi-step, cumulative trapping

Successive trapping of several QD@SiO_2 or pQD@SiO_2 clusters was sometimes observed. A typical PL intensity evolution for one of these processes is shown in Fig. S2. After a first cluster is trapped (t = 0) and meanwhile it is emitting, a second one reaches the trap, as evidenced by the abrupt intensity jump around 65 s. A third cluster was trapped at around 100 s without release of any of the previous ones. The three QD@SiO2 clusters then co-exist in the optical trap and co-generate the PL signal. From the overall temporal evolution of the PL intensity measured from the optical trap, the individual CER-predicted dependences (see Main Text, eq (2)) for each cluster can be derived (curves I, II and III, corresponding to first, second and third trapping events, respectively).

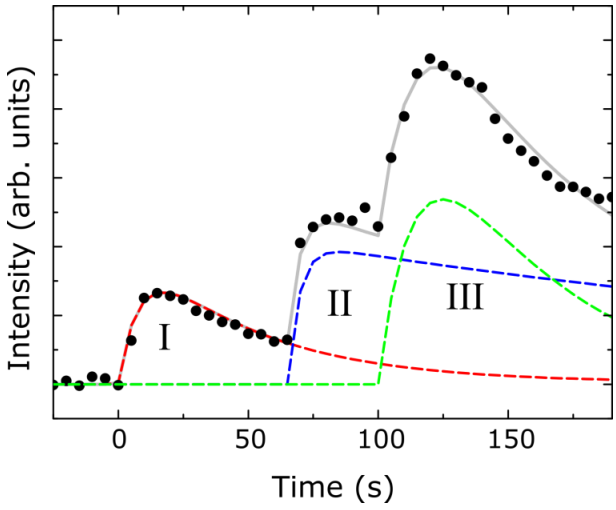

Figure S2. Temporal evolution of the intensity collected from the optical trap when three QD@SiO_2 clusters are emitting at the same time (black dots). The sum of three independent CER curves, starting at different times (I, II and III, dashed curves) fits the intensity profile observed (grey curve). Activation and decay rates for clusters I, II and III in this sample experiment are, respectively: k_a=*0.13±0.02* s^{-1}, k_d=*0.021±0.003* s^{-1}; k_a=*0.20±0.02* s^{-1}, k_d=*0.003±0.001* s^{-1} and k_a=*0.076±0.005* s^{-1}, k_d=*0.019±0.001* s^{-1}.

Supporting reference

(1) Banin, U.; Bruchez, M.; Alivisatos, a. P.; 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* (2), 1195–1201.