THE EFFECT OF SINGLE NANOCRYSTAL BLINKING ON THE LUMINESCENCE DECAY OF ENSEMBLES

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Nanocrystal (NC) systems commonly exhibit multiexponential photoluminescence (PL) decays. Though many explanations have been provided that hypothetically explain the manifestation of the stretched-exponential form, there has been little experimental evidence to indicate the success of one model over any other. Interestingly, the influence of single NC intermittence on the decay curves of NC ensembles has so far been overlooked. We suggest that the anomalous curve elongation is intimately liked to the power-law blinking statistics of single NCs.

Figure 1 pictorially summarizes a simple model used to investigate the effect that the single NC blinking phenomenon has on the anomalous multiexponential behaviour of NC ensembles. Using stable distribution theory we derive a simple fitting form for the analysis of PL decay curves which allows the extraction of both intrinsic recombination rates linked to quantum confinement and power-law exponents typifying blinking statistics of single nanocrystals. This approach is then applied to the PL decay curves of a series of silicon NCs embedded in silicon oxide with average sizes within the range 1.5 to 4nms. These results are shown in figure 2a and b; and are compared with parameters extracted using the empirical stretched exponential law figures 2c and d.

A theoretical tendency of recombination rate with emission energy as determined by Delerue et. al. is plotted on figure 2b for comparison. Note that the extracted values represent a much better agreement with theory than the inverse stretched exponential lifetime which is consistently some order of magnitude higher. Remarkably the extracted α values also agree perfectly with the expected values of the blinking exponent which are generally determined experimentally within the range $1<\alpha_{on/off}<2$, and predicted theoretically to be exactly equal to 1.5 for normal diffusion and to fall within the range $1.5<\alpha_{on/off}<2.5$ where anomalous diffusion and reaction is applicable. These results represent a promising development as the the same technique can potentially be applied to the analysis of carrier recombination in a great number of other quantum confined systems and in addition, may provide a simple alternative for the extraction of nanocrystal blinking statistics under a much wider range of conditions than is possible in difficult to execute single nanocrystal spectroscopy experiments.

References:

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Figures:

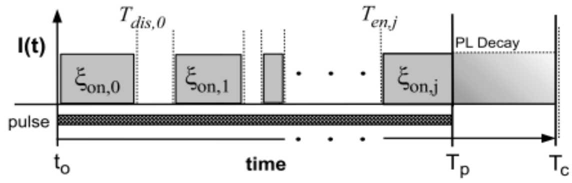

Figure 1: Events contributing to luminescence decay curves for blinking nanocrystal ensembles

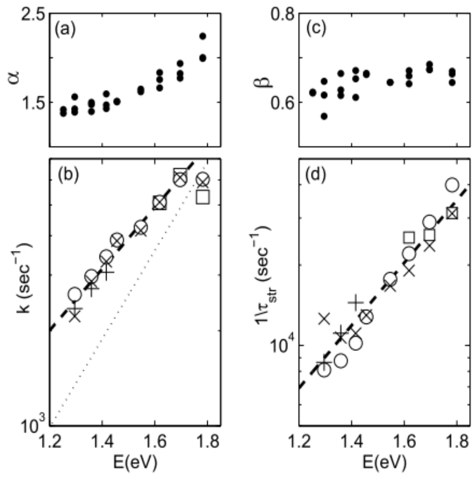

Figure 2: Recombination rates and associated parameters with observation energy resulting either from the derived blinking form (figures a and b) or the conventional stretched-exponential figure (c and d) for silicon nanocrystal samples.