Intermittency In Quantum Dots and Other Fluorophores

Abstract

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The purpose of the dissertation is to provide a theoretical explanation to the phenomenon of fluorescence intermittency. This is achieved by proposing a model of Multiple Recombination Centers (MRC), which is shown to successfully reproduce the main features of the phenomenon. Virtually all known types of optically active nanoscale objects to date show extremely long correlations in the fluctuations of fluorescence intensity. Experimentally collected intensity trajectories from such single fluorophores show fluctuations on the timescales longer than seconds. This phenomenon is generically referred to as fluorescence intermittency or blinking. In colloidal quantum dots blinking often assumes the shape of a random telegraph-like intermittency, a stochastic series of ``ON'' and ``OFF'' time intervals. Amazingly, the distribution of these ``ON'' and ``OFF'' times follow a universal power law dependence. Spectral characterization of trajectories effectively renders blinking an optical 1/f type noise. By a suitable Bayesian estimation method we point out that the conventional method of analysis using intensity histograms is problematic for such trajectories: The qualitative properties of the distributions strongly depend on the threshold value chosen for the separation the ``ON'' and ``OFF'' states. We propose the phenomenological MRC model for the quantum dot and its environment by modeling the multiple channels of non-radiative relaxation as a collection of a few interacting two-level systems. We show how this model of multiple recombination centers reproduces key experimental features of blinking, including the strong threshold dependence. After a survey of existing models of blinking, we show that the MRC model is the only self-consistent model that can explain the long-range correlations found between blinking times. Beyond quantum dots, a carefully performed spectral analysis of intensity fluctuations observed in other fluorophores such as self-assembled quantum dots, nanorods, nanowires, and some organic dyes, reveals the amazing similarity in the optical properties of these nanoscale systems. This similarity allows us to postulate a universal physical mechanism underlying the blinking phenomenon, based on the framework of the MRC model. Useful constrains are thus provided in the further efforts searching for the detailed microscopic mechanism of fluorescence intermittency.