An approach to analysis of optical spectra of colloidal quantum dots and their aggregates: taking account of inhomogeneous broadening, blinking, and FRET

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Förster resonance energy transfer (FRET) in aggregates of colloidal quantum dots with narrow size distribution manifests itself experimentally as a red-shift and quenching of the emission spectrum upon aggregation. A mathematical model has been developed to describe this process [1]. The input data for the model are the absorption and emission spectra of individual particles. However, typical experimental absorption and photoluminescence spectra of colloidal quantum dots are usually considerably inhomogeneously broadened and therefore cannot be directly used for calculations. Fortunately, the total broadening can be easily decomposed into homogeneous and inhomogeneous parts by using the fundamental Kennard–Stepanov–van Roosbroek–Shockley (KSvRS) relation between the absorption and emission spectra. Figure 1 illustrates this issue. As a result, given the experimental absorption and emission spectra of nonaggregated and aggregated particles, the model can be used to calculate the FRET efficiency and the fraction of particles in the OFF state (in the context of blinking).

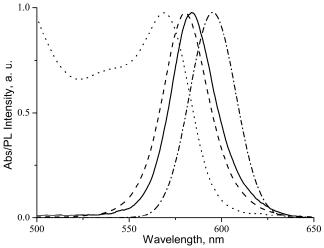


Figure 1 Experimental absorption (dotted curve) and emission spectra of nonaggregated (dashed line) and aggregated (solid curve) CdSe quantum dots with an average diameter of 3.5 nm [2]. The dash-dotted curve shows the emission spectrum of the ensemble of nonaggregated nanoparticles calculated from the KSvRS relation assuming a Gaussian shape of the spectrum and no inhomogeneous broadening. The deviation of this curve from the experimental one contains the information on the inhomogeneous broadening. The dash-dotted curve should also be the aggregate emission spectrum in the limit of infinitely fast FRET and no blinking. Its deviation from the solid line can be used to calculate the FRET efficiency.

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