

Exciton Dynamics in freely diffusing quantum dots in aqueous medium

Anindya Datta

Department of Chemistry, Indian Institute of Technology Bombay, Powai, Mumbai 400 076, India

e-mail: adutta@iitb.ac.in

Fluorescence Correlation Spectroscopy (FCS) and Fluorescence Lifetime Correlation Spectroscopy (FLCS) have been used to understand the dynamics of light-induced phenomena in two kinds of quantum dots (QDs) in aqueous solutions: Cu(I)-doped CdS (Cu:CdS) QDs [1] and CuInS₂ (CIS) QDs [2]. The Cu:CdS QDs exhibit photoactivation at higher excitation powers due to conversion of dim/dark particles to bright ones. Cu(I) doping is found to decrease the extent of dispersity in blinking kinetics, that is observed for undoped QDs due to the occurrence of trap states of various kinds. This is explained in the light of the predominance of radiative recombination involving holes captured by Cu(I) in the doped QDs. Significant suppression of Auger effect is another consequence of doping and consequent disruption of electron-hole correlation. On a different note, a ZnS shell is found to enhance the photoluminescence of CIS QDs remarkably. The initial correlation amplitude $G(0)$ increases with excitation power for $\lambda_{ex}=532$ nm, but decreases for $\lambda_{ex}=405$ nm, due to the difference in contributions from surface-assisted recombination for the two excitation wavelengths. Blinking times obtained from the Autocorrelation Functions (ACF) of the 100-200 ns lifetime component (core Cu-mediated recombination) are almost unaffected by shelling, but those from the ACF for the 10-30 ns lifetime (surface states) increase markedly. Absence of cross-correlation between the two recombinative states of bare CIS QDs and the emergence of an anticorrelation with the introduction of ZnS shell is observed, indicating the diffusive nature of the two states for CIS-ZnS. The diffusion is inhibited in base CIS QDs due to the preponderance of surface states.

References:

- [1] S. Das, G. Rana, F. Ali and A. Datta, *Nanoscale*. **15** (2023) 4469.
- [2] P. K. Singha, T. Kistwal and A. Datta, *J. Phys. Chem. Lett.* **322** (2023) 1073