

Spectral jitter of single CdSe/ZnS nanoparticles: Where is the charge?

D. Braam¹, A. Mölleken¹, G. M. Prinz¹, M. Geller¹ and A. Lorke¹

¹*Fakultät für Physik and CENIDE, Universität Duisburg-Essen, Lotharstraße 1, 47057 Duisburg, Germany*

Blinking and spectral jitter are both limiting the optical properties of semiconductor nanoparticles. They are generally attributed to a charge diffusion in the vicinity of the nanoparticle [1]. However, the position of this meandering charge is under strong debate. We use here time-resolved μ -photoluminescence (PL) of single CdSe/ZnS nanoparticles in combination with a simple model to locate the diffusing charge and find that it is trapped in the ligand layer.

PL spectra were taken at room temperature and fitted to a Lorentzian function to obtain both the emission peak energy and the corresponding linewidth (FWHM), two quantities which are correlated: A shift in the emission line to lower energies (a redshift) is accompanied by a broadening of the linewidth (see fig. 1 left inset). This is attributed to the Stark shift, by which the electric field of an outside charge carrier influences the nanoparticle's electronic structure and shifts the emission energy to the red. Due to the quadratic nature of the Stark effect, a perturbation at strong fields is more effective than at small fields, thus inducing a stronger broadening at higher field strengths.

Plotting the redshift over the FWHM, a superlinear correlation becomes apparent (see fig. 1). Employing a simple charge carrier model, the appropriate fit yields a mean position fluctuation of $\delta r = 0.8$ nm. Furthermore, it is possible to determine the distance between charge carrier and nanoparticle core. We find that most of the data lies in the range between 3 and 5 nm (see fig. 1 right inset). Because the particles consist of about 2 nm core, 1 nm shell and 2 nm ligands, we conclude that the diffusing charge carrier is predominantly located within the ligand layer.

Measurements of nanoparticles under the influence of photooxidation support these findings.

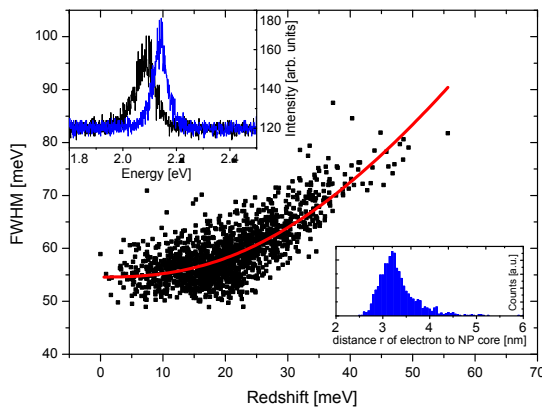


Figure 1: Linewidth over redshift showing a superlinear behaviour. The insets show exemplarily two representative spectra (left) and the spatial distribution of the diffusing charge with respect to the NP core (right).

[1] S. A. Empedocles, M. G. Bawendi, *Science* **278**, 2114 (1997).