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Spectroscopic signature of shape anisotropy in single CdSe nanocrystals

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The understanding of the photophysical properties of semiconductor nanocrystals is of utmost importance for the development of their applications in the fields of laser technology, biological labeling, nano-electronics and quantum information. Single nanocrystal spectroscopy and the theoretical model developed by Efros, Rosen et al. [1] have provided a deeper understanding of the properties of the band edge exciton fine structure in spherical CdSe NCs with ellipsoidal perturbations.

We study single zinc blende CdSe nanocrystals for the high symmetry of their cubic structure, which makes them very sensitive to deformations around the spherical shape [2]. We have developed a photoluminescence excitation technique (PLE) to probe the whole band edge exciton fine structure. We observe both split 1^L and 1^U states, which we attribute to shape anisotropy. The polarizations of the doublets are anticorrelated as predicted by the theory [3]. The split states are not orthogonally polarized, which can be interpreted as the result of non-orthogonal perturbations of the spherical shape.

Moreover, states coupling induced by an external magnetic field gives oscillator strength to forbidden transitions, allowing to resolve for the first time all the eight states of the band edge exciton fine structure, as shown in **Fig. 1** for an elongated nanocrystal. Optical spectroscopy therefore produces the full spectral fingerprint of single nanocrystals, which is essential to test long standing theoretical models.

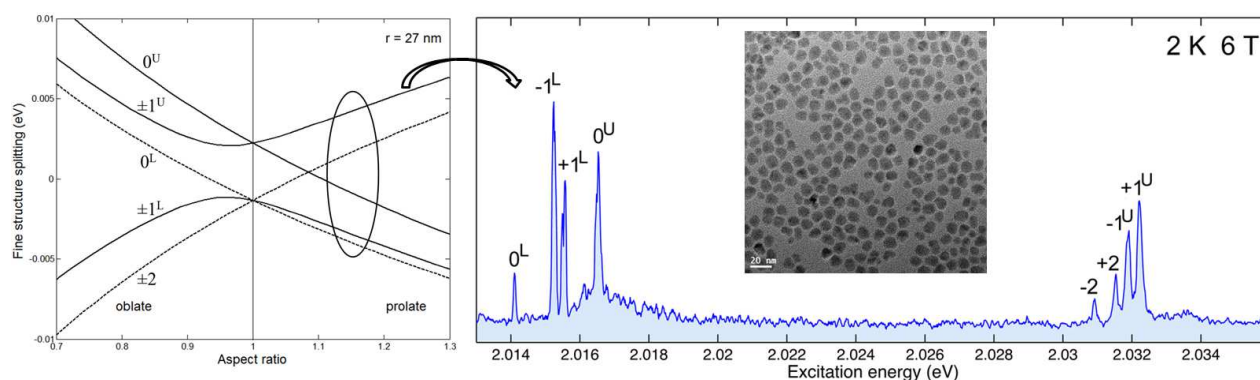


Fig. 1 Left : band edge exciton fine structure splitting as a function of the aspect ratio for zinc blende CdSe nanocrystals (dotted lines correspond to dark states, solid lines indicate bright states). Right : PLE scan at 2 Kelvin and 6 Tesla showing the whole fine structure of the band edge exciton for a single elongated nanocrystal.

References

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