LAPHIA axis : Emerging photonics and Materials

Spectroscopic signature of the entire band-edge exciton fine structure in single CdSe nanocrystals

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The understanding of the optical and electronic properties of semiconductor nanocrystals (NCs) is of extreme 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 size dependent emission properties of the band-edge exciton in CdSe NCs ranging from spherical to ellipsoidal shape.

Effects of shape anisotropy in colloidal CdSe nanocrystals have recently been observed, consisting in orthogonal linearly polarized bright state spectral doublets [2]. Two effective mass models were developed to explain the source of the anisotropy-induced lift of bright states degeneracy, including respectively anisotropic exchange interaction [2] and valence band mixing, which is the effect of shape anisotropy on the valence band energies [3]. The two models differ in the predictions of the spectroscopic characteristics of the lower and upper bright states, ($\pm 1^{L,U}$). Since the $\pm 1^{U}$ state is not observable in photoluminescence, due to its fast relaxation to the lower states manifold, these models have not been fully tested yet.

We study single spheroidal CdSe nanocrystals having a zinc blende crystal structure. Since there is no crystal field, the heavy hole and light hole bands are degenerate for spherical shape, making the band-edge exciton fine structure very sensitive to shape deformations. The study of photoluminescence at low temperature and under magnetic field enables us to establish a relationship between the spectroscopic fingerprint of the band-edge exciton fine structure and nanocrystal morphology according to the existing models [4].

In particular, we find that nanocrystals exhibiting anisotropy-induced bright state splittings have a prolate shape. These splittings, which spread up to ~ 1 meV, can be tailored by applying a magnetic field oriented in the plane perpendicular to the axis of the main deformation, eventually restoring the bright state degeneracy. This fine-tuning of individual quantum states energies can be helpful for quantum technological applications, such as the generation of polarization entangled photon pairs.

Furthermore, we develop a photoluminescence excitation technique (PLE) to probe the entire band-edge exciton fine structure. We observe an upper bright state splitting comparable to that of the lower bright state and anticorrelated polarizations between the split components of

$\pm 1^L$ and $\pm 1^U$ states. These results show that the bright states splittings are mainly due to valence band mixing [3].

Finally, combining shape anisotropy-induced splitting with an applied magnetic field, which produces bright – dark states coupling and Zeeman splitting, we reveal for the first time the entire 8-state band-edge exciton fine structure on a single nanocrystal, which is shown in Figure 1 [5]. This result corroborates the state ordering predicted by long standing theoretical models and provides a rigorous spectroscopic example for comparing with any future models.

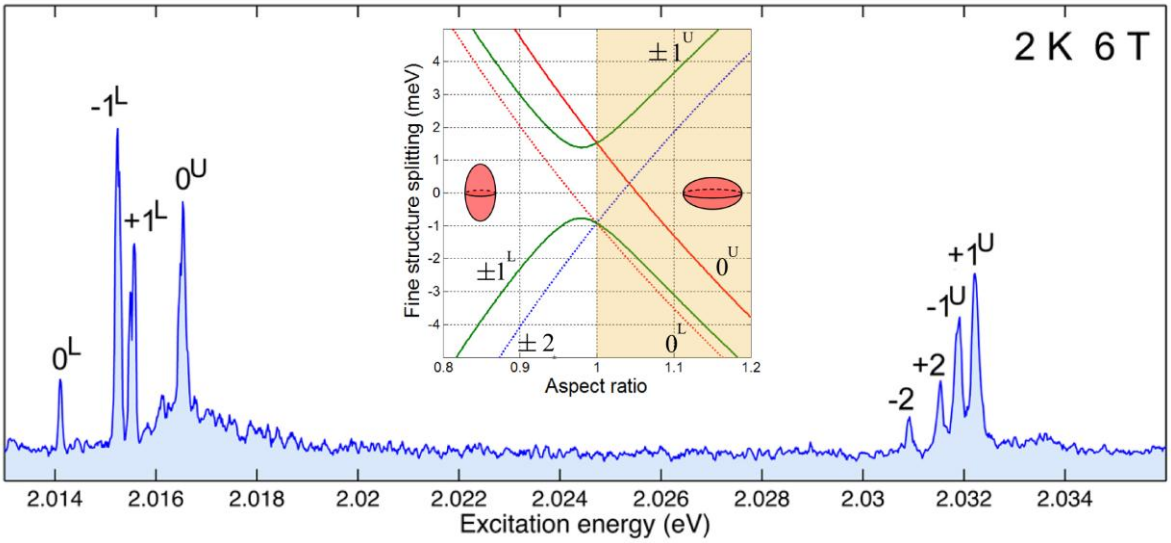

Figure 1 : PLE scan at 2 Kelvin and 6 Tesla showing the entire 8-state fine structure of the band-edge exciton for a prolate nanocrystal. Inset : band-edge exciton fine structure splitting as a function of the aspect ratio for zinc blende CdSe nanocrystals (dotted lines correspond to dark states, solid lines indicate bright states).

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