

Magneto-optical study to reveal the exciton fine-structure in InP/ZnSe core/shell quantum dots

Annalisa Brodu¹, Mariana Ballottin², Dorian Dupont³, Mickael Tessier³, Zeger Hens³, Peter C. M. Christianen², Celso de Mello Donega¹, and Daniel Vanmaekelbergh¹

¹Debye Institute for Nanomaterials Science, Utrecht University, The Netherlands

²High Field Magnet Laboratory, IMM, Radboud University, The Netherlands

³Physics and Chemistry of Nanostructures, Ghent University, Belgium

Colloidal semiconductor nanocrystals or quantum dots (QDs) are of significant interest in nanoscience and optoelectronic applications due to their size-tunable emission spectrum in combination with broad absorption and excitation spectra. For decades, Cd-chalcogenide QDs have been the workhorse in this field and have reached a very mature level. However, the implementation of QDs in commercial devices requires toxicologically harmless materials. As a result, the demand for Cd-free colloidal QDs with similar optical performance as Cd-chalcogenide QDs is rising rapidly. In this respect, the development of III-V QDs, especially of InP, has received increasing attention in recent years. Therefore, knowledge of the electronic structure of InP-based core/shell QDs is of large interest.

A powerful method to understand the electronic states and unravel the exciton fine-structure of QDs is to perform spectroscopic studies under high magnetic fields and at low temperatures [1-3]. For this reason, in this work [4] we studied the exciton fine-structure of InP/ZnSe QDs with various core diameters using different techniques: fluorescence line-narrowing (FLN) spectroscopy (Fig. 1a), polarized photoluminescence (PL) spectroscopy, and time-resolved photoluminescence (TRPL) spectroscopy, under high magnetic fields up to 30 T at temperatures down to 4 K. The high magnetic field allowed us to obtain a considerable Zeeman splitting of spin degenerate states and to induce significant mixing of exciton levels with different angular momentum. Combined with detection of the polarization of the emitted photons, the nature of the states has been resolved unambiguously: the (S_e, S_h) exciton state consist of a (± 2) optically passive state, followed by a (± 1) optically active state, followed by several other states with different angular momentum. The energy difference between the optically active and passive state is between 5-15 meV, increasing with decreasing QD size (Fig. 1b).

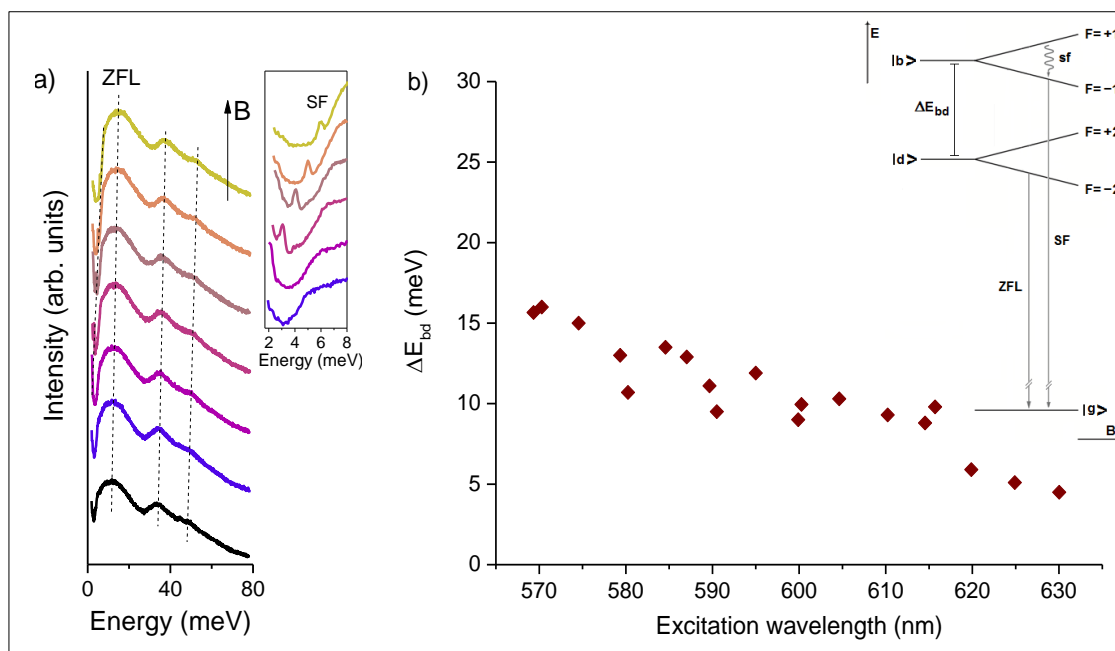


Fig. 1 InP/ZnSe core/shell QD sample: **a)** FLN spectra at fields from 0T to 30T. The Zero Phonon Line (ZFL) is highlighted. Insert: Zoom in of the low energy that shows the spin flip (SF) due to the Zeeman effect. The resonantly excited state is set to 0meV. The spectra are vertically shifted for clarity. **b)** Energy difference (ΔE_{bd}) between the optically active (b) and passive (d) state as a function of FLN excitation wavelength (exciton energy for given QD size). Insert: Energy level scheme used to interpret the data. It is composed of a ground state (g) and two excited state (b,d). The Zeeman splitting of the states with projection of total angular momentum $F=\pm 1$ (state b) and $F=\pm 2$ (state d) is also shown.

- 1) F. J. P. Wijnen *et al.*, Phys. Chem. B, **2008**, 78, 235318
- 2) J. H. Blokland *et al.*, Phys. Chem. B, **2011**, 83, 35304
- 3) A. Granados del Águila *et al.*, ACS Nano, **2014**, 8 (6), 5921
- 4) A. Brodu, MS in preparation