Indirect excitons in colloidal core-crown nanoplatelets

Among the II-VI semiconductor heterostructures at the heart of optoelectronic and spintronic applications, one finds the colloidal core-crown nanoplatelets whose thickness is controlled at the level of the atomic monolayer. Synthesized for the first time in 2014, they arouse a growing interest due to their relatively easy synthesis and their remarkable optical and electronic properties closely related to the generation of indirect excitons. These latter are photo-created with an electron in the core (CdSe) and a hole in the crown (CdTe), which leads to a poor overlap of the wave functions of both particles and to a long life of these excitations. Studies of the fine structure and spin physics of such indirect excitons are essential steps in the race for applications, and have not yet been addressed. This is why the Phocos team at INSP in collaboration with the LPEM of the ESPCI, the Universities of Cambridge, Linköping, Radboud and Sydney explored these topics through polarization and time-resolved luminescence experiments as a function of the temperature and under strong magnetic field, and showed the strong applicative potential of such colloidal heterostructures CdSe/CdTe.

The linear polarization of the emission of sets of nanoplaquettes after linearly polarized resonant excitation was jointly analyzed with the circular polarization rate under resonant circular excitation and as a function of a magnetic field up to 30 teslas [Figs. (a) and (b)]. The modeling of all the results allowed us (i) to conclude the existence of two bright states with crossed linear polarizations, (ii) separated by about 50 µeV, (iii) to measure the Landé factor of excitons [Fig. (c)]. Pump-probe quantum beat spectroscopy experiments confirm the value of fine-structure splitting of bright states and lead to an estimate of the spin coherence time of several hundred picoseconds.

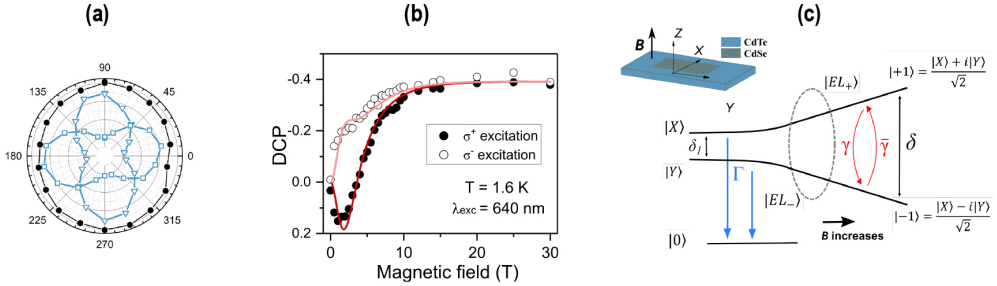

Figure 1
(a) Polarization diagrams of ensembles of resonantly excited nanoplatelets under zero magnetic field for two orthogonal incident linear polarizations (in blue) or non-resonantly (in black). (b) Circular polarization rate (Iσ + − Iσ -)/(Iσ + + Iσ -) of the emission after resonant circular excitations. The red lines represent adjustments of the experimental data by a model taking into account (i) the existence of linear emitter states separated by δ1 by a few tens of µeV in zero field, (ii) their «elliptisation» as and as the field increases, (iii) the measurement of the spin dynamics of excitons with field. (c) Schematic representation of the experimental configuration and levels of the fine structure of the indirect exciton.

From the time-resolved measurement of the circular polarization rate, we have also obtained the order of magnitude, in the microsecond domain, of the spin relaxation time between bright states. The whole results and their analysis clearly show the essential play of the equilibrium between the radiative recombination of the excitons and the relaxation of their spin.

In conclusion, the long lifetime and coherence time of the excitonic spin measured in core-crown CdSe/CdTe nanoplatelets demonstrate the ability of these systems to retain spin information, a fundamental aspect in spintronics.

Reference
Fine Structure and Spin Dynamics of Linearly Polarized Indirect Excitons in Two-Dimensional CdSe/CdTe Colloidal Heterostructures
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ACS Nano, 13, 10140 (2019).

Contact
Laurent Legrand: legrand@insp.jussieu.fr