

Spins in Colloidal Nanocrystals

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In this lecture spin related phenomena in colloidal nanocrystals (NCs) will be presented, including spin structure of neutral and charged excitons (trions) and spin dynamics of the excitons and carriers. Introduction to the optical properties of the colloidal nanocrystals will be given in their comparison with epitaxially grown quantum dots. Various types of colloidal nanocrystals will be considered: core only, core-shell [1-2], dot-in-rod [4-5], platelets [3] based on II-VI semiconductors, mainly CdSe/CdS. Specifics of the exciton energy structure and its influence on the recombination dynamics at different temperatures and strong magnetic fields will be considered with illustrating by experimental results.

Main part of the lecture will be devoted to spin physics in colloidal nanocrystals. Here we report on experimental and theoretical studies of the trion and exciton spin dynamics in core/thick-shell CdSe/CdS NCs. We have shown recently that photo-excitation of core/shell CdSe/CdS nanocrystals (NCs), which shell thickness exceeds 4 nm, leads to a single electron charging of NCs [1,2]. Time-resolved photoluminescence measurements were performed at low temperatures and in high magnetic fields up to 15 Tesla. From the decay of the photoluminescence intensity the trion radiative time of 8 ns was measured. It is independent of the magnetic field reflecting the fact that the trion ground state is always optically bright (i.e. allowed in electric-dipole approximation). This is in strong contrast to the exciton states in NCs which dynamics is controlled by a competition of the bright and dark states, which can be mixed either by magnetic fields or thermally.

Spin relaxation time of excitons is shorter than a nanosecond and are limited by time-resolution of the used setup. While for the trions it is very long up to 60 ns and it decreases by about two orders of magnitude down to 1 ns in strong magnetic field of 15 Tesla

Theoretical description of the polarization dynamics is complicated by the fact that we study an ensemble of CdSe/CdS nanocrystals with random orientation of their hexagonal axes to the magnetic field direction. The trion Zeeman splitting is controlled solely by the hole g-factor, which is strongly anisotropic: it is maximal for NCs oriented along magnetic field and zero for NCs oriented perpendicular to the field. However, the magnetic field mixing of the hole states, which accelerates spin relaxation in trion, is most efficient for the perpendicular orientation. The developed model approach accounts for all these conditions.

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