

# Spectroscopy of CdSe/ZnSe quantum dots with individual Fe<sup>2+</sup> ions

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## ABSTRACT

This work is devoted to the spectroscopic studies of self-organized CdSe/ZnSe quantum dots doped with individual Fe<sup>2+</sup> ions. Such dots belong to a wide class of quantum dots containing single transition metal ions. The main advantage of these systems consists in a possibility of all-optical control of the spin of a magnetic impurity interacting with the semiconductor lattice, but being isolated from any other magnetic ions located in a macroscopic sample. The results presented in this dissertation uncover a new, yet unexplored opportunity offered by such quantum dots, that is the possibility to tune and, eventually, to completely alter the magnetic properties of a single dopant. Such properties are mainly governed by the ground state of the dopant, which in the case of the Fe<sup>2+</sup> ion embedded in a bulk II-VI semiconductor is a singlet, nondegenerate state. We have demonstrated that this physical picture becomes qualitatively modified after placing the Fe<sup>2+</sup> ion in an epitaxial quantum dot, in which the ion ground state turns out to be nearly doubly degenerate, consisting of states corresponding to the ion spin projections onto the quantum dot growth axis of  $\pm 2$ . This finding has been evidenced both theoretically and experimentally.

The theoretical investigation has been carried out within the frame of a relatively simple point charge model describing the influence of the crystal environment on the properties of a magnetic dopant. Based on the model calculations, we have shown that the underlying reason for the qualitative change of the Fe<sup>2+</sup> ground state character is a strong structural strain of the quantum dot related to the lattice mismatch between the dot and barrier materials. Such strain alters the spectrum of the ion orbital states, which in turn induces distinctive changes in the ordering of the ion spin levels due to the spin-orbit and spin-spin couplings.

The predictions of the developed theoretical model regarding the energy structure of the Fe<sup>2+</sup> ion states have been confirmed by extensive magneto-optical experiments carried out on single CdSe/ZnSe quantum dots containing individual Fe<sup>2+</sup> ions. Based

on the results of such experiments, performed on over thirty randomly selected dots, we have established a general pattern of the photoluminescence spectra of various excitonic complexes coupled to the  $\text{Fe}^{2+}$  spin via the  $s,p-d$  exchange interaction. We have also closely examined the way in which these spectra evolve upon application of the magnetic field in both Faraday and Voigt geometries. All presented experimental results have been precisely reproduced by a spin Hamiltonian model. On this basis, we have determined typical values of the parameters describing the structure of the low-energy  $\text{Fe}^{2+}$  ion states in the studied dots as well as the character and strength of the exchange interaction between the ion and each confined carrier: the electron and the hole.