Optical probing of a Ni²⁺ spin in a charged or a neutral quantum dot: influence of local strain

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The spin of a carrier or of a single impurity in a semiconductor host can act as a bit of quantum information. In direct bandgap semiconductors, the spins of confined carriers can be optically controlled providing a natural spin-photon interface. This is also the case for some defect-localized spins, but this depends on a particular level configuration and optical selection rules. For non-optically active impurities the optical or electrical properties of the semiconductor can be used to interact with localized spins. In particular, confined carriers in quantum dots (QDs) can be exchange coupled with an embedded magnetic element and exploited to control its spin.

We report here on the optical properties of QDs containing a Ni^{2+} ion (3 d^8 , S=1, L=3, I=0) interacting with a charged or a neutral exciton (see Figure 1). The analysis of the magneto-optic properties of Ni^{2+} -doped self-assembled QDs shows that the spin structure of the magnetic atom is strongly influenced by the orientation of the strain distribution at the magnetic atom location. At zero magnetic field, low symmetry strain can mix the spin states of the atom increasing the number of possible optical transitions.

In charged Ni²⁺-doped QDs, up to nine emission lines can be observed. They correspond to all possible transitions between the three spin states of Ni²⁺ interacting with a single electron or hole in the excited and ground states of the QD, respectively. We identify optical transitions that share a common excited state. They form a series of Λ levels systems that can be optically addressed individually. This allows us to determine the energy levels structure in the ground and in the excited state of the charged QD. We measure the electron-Ni²⁺ and hole-Ni²⁺ exchange interaction and show that the exchange interaction with the hole is anti-ferromagnetic and at least ten times larger than for the electron. For neutral QDs, the low symmetry of the dots allows to observe both the bright and the dark excitons and to follow their evolution under magnetic field. A spin-effective model that includes the local anisotropy of the strain experienced by the magnetic atom agrees with the main features of the experimental observations. Low symmetry terms in the hole-Ni²⁺ exchange interaction have to be taken into account for a detailed description of the emission spectra.

The identification of an optical Λ level structure in the charged Ni²⁺-doped QDs opens up some possibilities for the manipulation of the Ni²⁺ spin under resonant optical excitation.

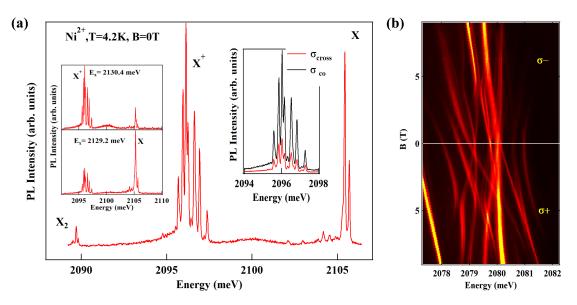


Figure 1: (a) Photoluminescence (PL) of the exciton, charged exciton and biexciton in a Ni^{2+} doped QD. Left inset: PL for 2 different excitation energies. Right inset: Circularly polarized PL of the charged exciton for a resonant excitation on an excited state. (b) Intensity map of the longitudinal magnetic field dependence of the PL of a positively charged Ni^{2+} -doped QD.

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