Magnetic Ground State of an Individual Fe^{2+} Ion in a Strained Semiconductor Quantum Dot

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Spin manipulation of individual impurities has attracted a lot of research attention over the last years [1-3]. Important contribution to this field can be done using quantum dots (QDs) containing various single transition metal ions, as was recently shown by the measurements of Mn^{2+} and Co^{2+} spin relaxation dynamics in different QD systems [2] or by our observation of coherent precession of a single Mn^{2+} spin [3].

One of the strongest motivations for the research in this area is a possibility to obtain long spin coherence time of an individual magnetic moment. From that perspective the nuclear-spin-free Fe^{2+} ion embedded in a QD seems a promising system. However, it was not considered as a candidate for quantum information applications, since the Fe^{2+} ion in bulk zinc-blende or wurtzite II-VI semiconductors was found to inherently exhibit a single non-degenerate ground state and thus regarded as not able to store any quantum information.

In this work we demonstrate that by using the strain of a semiconductor QD it is possible to tailor the energy spectrum of the Fe^{2+} ion to exhibit doubly degenerate (i.e., magnetic) ground state. Moreover, this ground state is composed of states corresponding to the ion spin projections $S_z = \pm 2$, which makes those two states less prone to decoherence, e.g., by residual in-plane magnetic field. Our concept is evidenced both theoretically and experimentally. From the theoretical side, we find that strong structural strain of the QD alters the spectrum of the ion orbital states, which in turn induces a distinctive changes in the ordering of the ion spin levels due to the spin-orbit coupling. The experimental proof is based on the results of photoluminescence (PL) studies of a novel QD system: self-assembled CdSe/ZnSe quantum dots doped with individual Fe^{2+} ions. A direct fingerprint of a nonzero spin of the Fe^{2+} ion ground state is a pronounced twofold splitting of the emission lines visible in a QD PL spectrum, which is observed for all three excitonic complexes (Fig. 1). In each case, the splitting originates from the *s,p-d* exchange interaction between the ion and confined carriers, which leads to two different energies of the optical transitions depending on the spin projection of the Fe^{2+} ion. Our analysis is complemented by the measurements of a QD PL spectrum evolution in magnetic field, which allow us to determine the character and strength of the *s,p-d* exchange and to obtain the ion g-factor of 2.0, exactly as expected for the Fe^{2+}.

An excellent agreement between our model and experimental results unequivocally confirms the strain-induced magnetic character of a single Fe^{2+} ion in a CdSe/ZnSe QD. Such a novel QD system is thus a prominent candidate for quantum information processing, since both the CdSe lattice and Fe^{2+} ion can be free of any nuclear spin fluctuations.

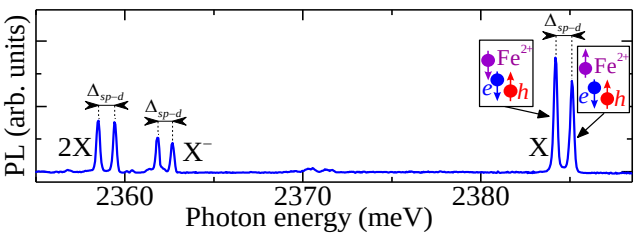

Fig 1. PL spectrum of a CdSe/ZnSe QD with a single Fe^{2+} ion ($\Delta_{sp\text{-}d}$ represents the *s,p-d* exchange splitting visible for all excitonic complexes).

[1] C. Yin *et al.*, *Nature* **497**, 91 (2013).
[2] J. Kobak, T. Smoleński *et al.*, *Nat. Commun.* **5**, 3191 (2014).
[3] M. Goryca *et al.*, *Phys. Rev. Lett.* **113**, 227202 (2014).