Coherent oscillations of a single Mn²⁺ spin in a CdTe quantum dot

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Semiconductor quantum dot (QDs) containing single magnetic impurity is a new form of diluted magnetic semiconductors which has recently attracted a lot of interest [1,2]. Not only is it a model system for investigation of many spin-related phenomena, but also a promising candidate for the building block of future information storage devices and a tool for quantum computation [3]. Particularly, the possibility to read and manipulate the electronic spin state of the magnetic ion has been shown in the case of CdTe quantum dots with a single isoelectronic Mn²⁺ impurity [4,5]. However, those experiments concerned only non-coherent phenomena, while application in quantum computing requires the ability to coherently manipulate and process the spin. Coherent measurements, including observation of Rabi oscillations and long coherence time, were performed on an ensemble of colloidal ZnO QDs containing many Mn²⁺ dopants [6]. Here we present a direct observation of coherent oscillations of a single Mn²⁺ spin embedded in a CdTe QD placed in magnetic field.

The sample used in the experiment contains a single layer of self-assembled CdTe/ZnTe QDs. The dots contain a low amount of Mn²⁺ ions, so that selection of single dots with exactly one magnetic ion is possible. In order to probe the spin state of the single Mn²⁺ impurity we performed a time-resolved measurement of the absorption of a QD containing such an ion. The QD was resonantly excited with two circularly polarized picosecond laser pulses. The energy of the photons was tuned to the transition energy of an exciton-Mn²⁺ complex with arbitrary chosen spin state of the magnetic ion. The absorption was detected by using the excitation transfer to a neighboring QD [5,7] and observation of the emission from this dot. The presence of a second, coupled QD not only enabled the detection of the absorption, it also assured a very short exciton lifetime in the absorbing QD, crucial for the temporal resolution of the experiment. The delay between the two pulses was precisely controlled so that the second pulse could probe the evolution of the investigated system after the first pulse.

The system under investigation was placed in a magnetic field applied in the Voigt configuration, parallel to the surface of the sample. Under such conditions the eigenstates of the Mn^{2+} ion depend on whether the exciton is present in the QD. Upon absorption of the first laser pulse the exciton- Mn^{2+} complex is created in a specific spin state, defined by the photon energy. However, when the exciton tunnels to the neighboring QD, the Mn^{2+} ion is no longer in the eigenstate due to the presence of magnetic field. Therefore the spin state of the magnetic impurity starts to oscillate and the probability of the absorption of the second pulse (of the same energy as the first one) depends on the delay between pulses. By observing the luminescence intensity of the neighboring QD we are able to reproduce those oscillations. We can also determine the dephasing time $(\mathrm{T_2}^*)$ of the Mn^{2+} ion, limited mainly by the hyperfine interaction and the crystal field originating from the strain of the QD material.

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