Optical control of a Mn spin embedded in a quantum dot

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Abstract. We demonstrate a high degree of an optical spin preparation of a single Mn atom embedded in a CdTe/ZnTe quantum dot (QD). Due to the strong exchange interaction of the manganese atom with an exciton injected into the QD the spin orientation can be achieved by quasi-resonant or fully-resonant optical creation of the polarized electron-hole pairs. A measured spin memory of the isolated Mn atom, in most of the cases, is in the microsecond range, and depends on the built-in strain in the quantum dot. During the resonant optical pumping process exciton spin-flip can occur without a change of the Mn spin providing a way to directly read the dynamics of the pumped spin state. The manganese spin orientation is achieved in a few tens of ns.

1. Introduction
Spintronics in semiconductors is very attractive as it combines advantages of semiconductor and magnetic materials. The new generation of devices will fulfil the demands in increased data processing, nonvolatility, decreased power consumption and increased integration densities. One of the good candidates for these devices are diluted magnetic semiconductors (DMS) [1], where the spin of magnetic atoms (Mn) are strongly coupled to spin polarized carriers introduced optically. It is known that in the absence of carriers and under a magnetic field, highly dilute ensemble of Mn present a spin relaxation time in the millisecond range [2]. However, the dynamics of such ensembles can be much faster at zero field [3].

In our work a single Mn atom embedded in a CdTe/ZnTe QD is used. The spin of the atom is controlled owing to the coupling with the exciton injected optically into the QD. We demonstrate the ability of a single spin orientation by optical pumping. The measured spin memory is in a microsecond range at zero magnetic field [4].

2. Samples and experiment
The samples used in the experiments contain CdTe/ZnTe QDs doped with a single Mn atom. Growth and optical addressing of such QDs were achieved recently [5, 6, 7]. The static properties of these systems are now well understood: when a Mn atom is included in a II-VI QD, the spin of the optically created electron-hole pair interacts with the five $d$ electrons of the Mn (total spin $S = 5/2$). This leads to a splitting of a pure QD PL spectrum into six $(2S+1)$ components, as shown in Fig. 1. This splitting results from the spin structure of the confined holes which
are quantized along the QD’s growth axis with their spin component taking only the values $J_z = \pm 3/2$ [8]. The PL energy is determined by the relative projection of the Mn and hole spins [9]. The intensity of each PL line reflects the probability for the Mn to be in one of its spin states and is probed at the moment the exciton recombines [10]. As the Mn spin fluctuates during a time averaged PL measurement, the six lines are observed simultaneously.

To optically pump the Mn spin, Mn-doped QDs were quasi-resonantly excited with a tunable continuous wave (CW) dye laser. To record the spin transients, the linear polarization of the excitation laser was modulated between two orthogonal states by switching an electro-optic modulator with a rise time of 5 ns, and converted to circular polarization with a quarter-wave plate. Trains of resonant light with variable duration were generated from the CW laser using acousto-optical modulator with a switching time of 10 ns. The circularly polarized collected light was dispersed by a 1 m double monochromator before being detected by a fast avalanche photodiode in conjunction with a time correlated photon counting unit with an overall time resolution of $\approx 40$ ps.

### 3. Optical orientation of the Mn spin state

Under the non-resonant optical injection of unpolarized or spin polarized exciton the Mn spin is oriented in the exchange field of the confined electron-hole pair. In this way, the Mn spin population becomes non-equilibrium. For example, under a continuous injection of $\sigma^+$ excitons, the probability of finding a Mn spin in $S_z = +5/2$ state decreases with time, while $S_z = -5/2$ increases.

Fig. 2 and 3 summarize the main features of a time-resolved optical orientation experiment performed under quasi-resonant excitation (into the first excited state of a QD). In the experiment a QD is sequentially excited by trains of circularly, oppositely polarized trains of light of small intensity. A time-resolved PL is detected in circular polarization from the high energy peak (e.g., $\sigma^-$ polarization detects the $S_z = -5/2$ spin population). Switching the circular polarization of the excitation we read a change of the PL intensity (Fig. 2a) which is with two transients: first one is reflecting an increase of the $S_z = -5/2$ population under $\sigma^+$ excitation, and the second demonstrate an efficient depletion of the same spin state under $\sigma^-$ excitation. In Fig. 2b we monitor the two opposite X-Mn energy states in $\sigma^-$ polarization (i.e., $S_z = +5/2$ and $S_z = -5/2$ states) during the $\sigma^+ / \sigma^-$ excitation switch. The graph is reflecting a redistribution of the population of two opposite Mn spin states under spin-selective excitation. Relaxation processes tend to anti-align the Mn spin with the X exchange field to reach a thermal equilibrium on the X-Mn levels [11]. The observation of a dynamic spin orientation of the Mn denotes that the spin relaxation time of the Mn alone $\tau_{Mn}$ is longer than the relaxation within
Figure 2. PL transients at different values of the excitation power. Inset: power dependence of the inverse response time $\tau_r$, taken at the $1/e$ point of the spin-related transient. (b) PL transients recorded in $\sigma^-$ polarization on the high ($S_z = -5/2$) and low ($S_z = +5/2$) energy line of the X-Mn complex. (c) Simplified level diagram of a Mn-doped QD, as a function of Mn spin (X-Mn: bright exciton-Mn).

Figure 3. (a) PL transients recorded on QD1 (corresponding PL presented in the inset) under the optical polarization sequence displayed at the bottom of the plot. The spin distribution prepared by optical pumping is conserved during $t_{dark} = 3.5 \mu s$. (b) PL transients recorded on QD2. The amplitude of the pumping signal is restored after $t_{dark} \sim 3 \mu s$. From the delay dependence of this amplitude we deduce a Mn relaxation time of $\approx 700$ ns.

the X-Mn complex $\tau_{X-Mn}$.

Having a method to prepare a Mn spin, we performed pump-probe experiments to observe how the Mn polarization is conserved (Fig. 3). We induce a non-equilibrium distribution of the Mn spin with a $\sigma^-$ pulse. The pump laser is then switched off, and switched on again after a dark time $t_{dark}$. The amplitude of the pumping transient after $t_{dark}$ depends on the Mn spin relaxation in the dark. For QD1 (Fig. 3a) no transients are observed after a dark time of 3.5 $\mu$s. This demonstrates that in the absence of charges fluctuations (i.e. neutral QD (see inset of Fig.3a)) the prepared Mn spin is conserved over a $\mu$s. For QD2 (Fig. 3b) the amplitude of the pumping signal is restored after $t_{dark} \sim 3 \mu s$. From the delay dependence of this amplitude we deduce a Mn relaxation time of $\approx 700$ns. These two examples show that the measured spin relaxation is not intrinsic to the Mn spin but depends on its local environment.

4. Resonant optical pumping

The direct resonant excitation into the ground state of the X-Mn system is predicted [11] to perform a more efficient optical pumping of the Mn spin. Photon absorption followed by a spin relaxation within the X-Mn complex will empty the Mn spin state which is resonantly excited.

To estimate the efficiency of this optical pumping process, we developed a two wavelength pump-probe experiment allowing an optical initialization and read-out of the Mn spin. In this experiment, a QD is resonantly excited in the high energy state of the X-Mn system by circularly polarized (i.e. $\sigma^+$) CW laser (pump)(inset of the Fig. 4). This decreases the population of the $S_z = +5/2$ state, and is reflected in the intensity of a time-resolved resonant PL measured at
the low energy of the X-Mn complex in $\sigma^-$ polarization. A decrease of about 75% of the PL is observed during the optical pumping process with a characteristic time of 70 ns. Observed PL decay is referred to the decrease of the absorption of the QD induced by optical pumping.

After each pump pulse we have to use a linearly polarized quasi-resonant excitation (probe) to restore the equilibrium distribution of the Mn spin (see Fig. 4). Otherwise the absorption of the resonant pulse stays minimized due to the Mn spin memory. Using quasi-resonant excitation we also probe the change of population of the $S_z = +5/2$ state induced by the resonant pulse.

![Figure 4. PL transients recorded on the low energy line of a Mn-doped QD (QD1) under the quasi-resonant (QD’s excited state) and resonant optical excitation sequence displayed at the bottom: (i) resonant PL produced by the pump when the probe is OFF, (ii) PL from the pump and the probe when the probe is ON and (iii) difference between the PL produced by the probe when the pump is OFF and when the pump is ON. Because of the Mn spin memory, no signature of pumping is observed when the probe is OFF (i). The optical pumping process is directly observed in the PL produced by the pump and latter on the PL from the probe laser. The inset presents the QD PL and the configuration of the resonant excitation and detection.](image-url)

5. Conclusions

Our results demonstrate an optical spin orientation of a single magnetic atom in a semiconductor QD at zero magnetic fields. High fidelity spin preparation is achieved by resonant excitation of the exciton-Mn system ground state. The new technique used here allows for an accurate and direct readout of the population of any X-Mn ground state.

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