All-optical spin switching in neutral or charged magnetic quantum dots

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Abstract. The spin dynamics in a single semiconductor quantum dot doped with a single Mn atom are analyzed. We consider a neutral and a negatively charged dot and in both cases we concentrate on the light hole-to-conduction band transition. Both electrons and light holes couple to the Mn spin via the strong exchange interaction. After the excitation by an ultra short laser pulse oscillatory spin dynamics take place, where electron or hole can flip their spin accompanied by a change of the Mn spin. The Mn spin dynamics can be controlled by excitation with additional pulses. Starting from a given initial state we demonstrate that the Mn spin can be flipped all-optically into a steady final state in both neutral or charged quantum dots.

1. Introduction

The control of a single spin opens up possibilities for effective quantum computing. While usually two-level systems are taken as basic building blocks (qubits) for quantum gates, recently it has been shown that quantum logic might be considerably simplified by using higher dimensional Hilbert spaces, i.e., systems with more than two levels [1]. A Mn atom has a spin of \( M = \frac{5}{2} \) due to its five electrons in the d-shell and thus six possible spin orientations \( M_z \). Recently quantum dots (QDs) containing a single Mn atom have been fabricated in II-VI (CdTe) [2] and III-V (InAs) materials [3]. The spectra of these dots exhibit a splitting of the exciton line into a set of six (CdTe) or five (InAs) lines, clearly indicating the pronounced exchange interaction between Mn and carrier spins. By applying a voltage in a Schottky gated structure a single carrier can be injected into the QD thus realizing a charged magnetic QD [4].

In this paper we will analyze possibilities to realize an all-optical switching of the Mn spin on picosecond time scales both in neutral and charged QDs. We consider a CdTe QD where Mn is an iso electronic impurity. Although the Mn spin is not directly accessible by laser light, a control of the Mn spin state can be achieved through the optical manipulation of the exciton because of the exchange coupling between these two subsystems. For a flat QD we have recently shown that by a suitable sequence of laser pulses, the Mn spin can be efficiently flipped from a given initial state into all other spin states [5]. In flat QDs the uppermost valence band state is typically of heavy hole (HH) type with a spin of \( S^h_z = \pm \frac{3}{2} \). If a HH exciton is created, only the electron can induce spin flips while the hole spin is pinned. This leads to a bottleneck in the spin flip dynamics, which can be overcome if in addition excitations of a light hole (LH) exciton are included [5]. Here we will show that an efficient Mn spin control is also possible if only LH...
excitons are excited. In fact, if the QD has a prolate shape or in the presence of a suitable strain field the uppermost VB state can be of LH type. Such a system has also been proposed to realize single-qubit operations in an optically driven QD [6].

2. Neutral dot

For a single CdTe QD doped with a single Mn atom we consider the lowest conduction band state ($S^c_2 = \pm 1/2$) and the uppermost VB state taken to be of LH type ($S^h_2 = \pm 1/2$). Both states are two-fold degenerate. Accordingly the electronic system consists of the ground state $|0\rangle$ without excitons, two bright exciton states $|L \pm 1\rangle$, two dark exciton states $|L \pm 0\rangle$ and the biexciton state $|LL0\rangle$. (The number in our notation is the angular momentum $S^c_2 + S^h_2$; the sign in $|L \pm 0\rangle$ refers to the sign of the hole spin.) The bright excitons are excited by circularly polarized, Gaussian laser pulses with a full width at half maximum of 100 fs. The coupling to the light is treated in the usual rotating wave and dipole approximation. A $\pi$-pulse creates one exciton in the system. The Heisenberg exchange interaction $H_{exc} = j_e \vec{M} \cdot \vec{S}^e + j_h \vec{M} \cdot \vec{S}^h + j_{eh} \vec{S}^e \cdot \vec{S}^h$ with the coupling constants $j_{e/h/eh}$ contains Ising type terms $\sim M_z S^c_z S^h_z$ and correlated spin flips terms $\sim \frac{1}{2} (M_+ S^c_+ S^h_+ + M_- S^c_- S^h_-)$ (same for electron-hole interaction). With our model we follow Ref. [7], which successfully explained the observed photoluminescence spectra of a Mn-doped QD. A scheme of the involved excitonic states including the couplings induced by the carrier-Mn and the carrier-light interaction is shown in Fig. 1(a). For this system the Liouville-von Neumann equation for the density matrix has been set up and solved numerically.

We start with the exciton system in the ground state and the Mn spin in the state with $M_z = -5/2$. Such a state can be prepared by applying a small magnetic field in $z$-direction. A $\sigma^+$ polarized laser pulse at $t = 0$ excites a $|L + 1\rangle$ exciton. Due to the exchange interaction both electron and hole spin can be decreased by one accompanied by a corresponding rise of the Mn spin by one. This manifests itself in exchange-induced Rabi oscillations, which are seen in the occupations of the bright and dark exciton states shown in Fig. 2. Before looking at the system including all interaction, we first show the single electron or hole flip. The electron-hole exchange interaction has been switched off in these cases. Figure 2(a) shows the flip of the electron, while the spin flip terms of the hole have been turned off. After the excitation the electron spin completely flips turning the bright exciton $|L + 1\rangle$ into the dark exciton $|L + 0\rangle$. Here, bright and dark exciton states are degenerate, such that the exchange-induced Rabi oscillations are resonant with a period of about 18 ps. When looking at the hole flip in Fig. 2(b) (here the electron spin is artificially pinned), it is seen that the oscillation is much faster with a period of about 2 ps and the hole spin flip turns the bright exciton $|L + 1\rangle$ to the dark exciton $|L - 0\rangle$. This oscillation is not resonant, i.e., its amplitude is only about 0.8, because bright and dark excitons are split by the Ising terms of the exchange interaction. Both oscillations couple only two states each and each raises the Mn spin by one or 0.8. If now both spin flips are allowed, the exciton can go from the bright exciton state $|L + 1\rangle$ via one of the dark excitons $|L \pm 0\rangle$ into the other bright exciton state $|L - 1\rangle$. This is shown in Fig. 2(c), where all exchange interactions (electron-Mn, hole-Mn and electron-hole) have been included. We find a rather complicated
behavior after the excitation at $t = 0$. First the dark exciton $|L − 0\rangle$ becomes occupied due to the fast hole spin flip. The occupation of the other dark exciton $|L + 0\rangle$ rises more slowly. From these states flips can occur into the other bright state $|L − 1\rangle$ resulting in a rather irregular oscillation of its occupation. Each oscillation is accompanied by a change of the Mn spin, which in principle could change by a maximum of two. However, with a single pulse the expectation value of the Mn spin reaches only about $−1.4$.

To change the properties of the oscillations and to increase the Mn spin flip $2\pi$-pulses can be used. Such a $2\pi$-pulse does not change the occupation of the exciton states, but it induces a phase jump by $\pi$ in the coherences between bright and dark exciton states [5]. Due to this phase jump the limits of the exchange-induced Rabi oscillation are shifted. This is exploited in the case of the laser sequence shown in Fig. 3(a), where the expectation value of the Mn spin $\langle M_z \rangle$ as well as the laser sequence are shown. The number of pulses and their arrival times have been randomly generated and the optimal sequence has been selected. After the excitation at $t = 0$ three $2\pi$-pulses are applied. The expectation value of the Mn spin rises in a non-monotonic way from $−5/2$ to $−1/2$. When it is close to $−1/2$ the exciton system is in the bright state $|L − 1\rangle$, i.e., both electron and hole spin have flipped. Now a $\sigma^−$ polarized pulse can be used to annihilate the exciton thus bringing the excitonic system back into its ground state. Since there is no exciton anymore, the Mn spin state is stable and does not change its value anymore. At any time later from this stable state again a flipping by two can be induced by a new sequence of $\sigma^+$ polarized $\pi$ and $2\pi$ laser pulse, here arbitrarily starting at $t=17$ ps. The expectation value of the Mn spin starts to rise to $+3/2$ after the excitation. This time only a single $2\pi$-pulse is sufficient to almost completely flip the Mn spin. When the expectation value is close to $+3/2$, a $\sigma^+$ polarized $\pi$-pulse drives the exciton system to its ground state. In this way the Mn spin can be changed by two or four, while excitons are only present in the system during the switching. These periods when excitons are in the system are indicated by green shaded areas in Fig. 3(a).

3. Charged dot

If a single electron is injected into the quantum dot the system changes profoundly. The exchange interaction now leads to a coupling between electron spin and Mn spin already in the ground state resulting in a twelfold ground state multiplet. The excited state contains an exciton and an additional electron, i.e., a trion with a fully occupied conduction band state. Since the LH also has a spin of $1/2$, the Mn-trion manifold consists also of twelve states. The trion can be excited by circularly polarized laser pulses and spin flips can take place in both ground state
Figure 3. Control of the expectation value of the Mn spin $\langle M_z \rangle$ (upper panel) with a given laser pulse sequence (lower panel) for (a) the neutral dot and (b) the charged dot. The green shaded areas indicate the presence of (a) excitons or (b) trions.

and trion state. A scheme of the electronic states, consisting of the electron states $|E \pm 1/2\rangle$ and the trion states $|T \pm 1/2\rangle$, as well as their couplings by light and exchange interactions is shown in Fig. 1(b). The ground state manifold has two states with well-defined Mn spin, either with both electron and Mn spin down $|E - 1/2, M_z = -5/2\rangle$ or up $|E + 1/2, M_z = +5/2\rangle$. A switching sequence for the Mn spin is shown in Fig. 3(b), where the expectation value of the Mn spin $\langle M_z \rangle$ and the laser sequence are shown. The green shaded areas mark the periods when a trion is present. Starting from the initial state $|E - 1/2, M_z = -5/2\rangle$, the trion is created by a $\sigma^+$ polarized $\pi$-pulse. In the trion state the LH spin can flip and raise the Mn spin by one. Again an additional 2$\pi$-pulse is used to complete the spin flip. Then, by a $\sigma^-$ polarized $\pi$-pulse the electronic system is driven back into the ground state, where now the electron can perform a spin flip rising the Mn spin to $-1/2$. This scheme is repeated, changing the Mn spin by two to $+3/2$. After another $\sigma^+$-excitation, the flip of the LH spin in the trion state changes the Mn spin to $+5/2$. When the trion is deexcited by a final $\sigma^-$ polarized pulse, the system is in state $|E + 1/2, M_z = +5/2\rangle$, which is again a stable spin state. Thus the Mn spin has been inverted.

4. Conclusions
In summary we have presented different scenarios for an all-optical control of the Mn spin in a single QD based on optical transitions between the LH band and the conduction band. Due to the strong exchange interaction correlated flips of electron and Mn spin or hole and Mn spin can take place. In the neutral dot, after creating a LH exciton by a $\pi$-pulse oscillations of the Mn spin set in which are accompanied by electron and hole spin flips. By applying 2$\pi$-pulses these oscillations can be efficiently controlled and the Mn spin can be flipped by two or four, while the carrier system returns to its ground state where no exciton is present. In a charged dot already in the ground state the electron spin is coupled to the Mn spin. In this case we have shown that by a sequence of $\pi$- and 2$\pi$-pulses the Mn spin can be inverted.

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