All-Optical Spin Manipulation of a Single Manganese Atom in a Quantum Dot

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For a CdTe quantum dot doped with a single Mn atom we analyze the dynamics of the Mn spin when the dot is excited by ultrashort laser pulses. Because of the exchange interaction with the Mn atom, electron and hole spins can flip and induce a change of the Mn spin. Including both heavy and light-hole excitons and using suitable pulse sequences, angular momentum can be transferred from the light to the Mn system while the exciton system returns to its ground state. We show that by a series of ultrashort laser pulses the Mn spin can be selectively driven into each of its six possible orientations on a picosecond timescale. By applying a magnetic field the total switching time and the required number of pulses can be strongly reduced.

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Diluted magnetic semiconductor quantum dots (QDs) are promising candidates for new applications in the field of spintronics such as devices based on an exciton-controlled magnetization [1,2] or optical switching devices [3]. If the QD is doped with a single magnetic impurity one enters the fascinating field of single spin manipulation [4] with possible applications in quantum information processing. Indeed, CdTe QDs doped with a single Mn atom can act as single photon emitters with an emission at six discrete frequencies [5] or they have been proposed for a qubit with six states [6]. Meanwhile, both CdTe [7] and InAs [8] QDs doped with a single Mn atom have been successfully fabricated and characterized by single dot photoluminescence (PL) spectroscopy. In particular CdTe QDs have been extensively studied in the past years [5,7,9,10]. Here the Mn acts as an isoelectronic impurity and the exchange interaction between exciton and Mn spin leads to a splitting of the exciton line at zero magnetic field into six lines.

The optical manipulation of the charge degrees of freedom in a single QD by ultrafast laser pulses has reached a high level of sophistication. Rabi oscillations have been observed on the lowest [11,12] and the excited exciton [13] transition as well as on the biexciton transition [14]. To make use of the spin states of the Mn atom in a QD it is essential to be able to selectively prepare these states. However, they cannot directly be addressed optically. Here we will show that nevertheless by suitable sequences of ultrashort optical pulses the spin of a single Mn atom in a single QD can be selectively driven into each of its six eigenstates. The basic mechanism is the exchange interaction between carriers and Mn spin giving rise to correlated flips of exciton and Mn spin. Combined with the excitation and deexcitation of the QD by circularly polarized pulses, angular momentum can thus be transferred from the light to the Mn atom. Our goal has been to create a physically transparent protocol where each individual step can be well interpreted. In the following we will first present this protocol and then explain in detail the individual steps.

For our calculations we follow the structural and electronic model that has been successfully used to describe the experimentally observed PL spectra [15]. We consider one electron and two hole states, the lowest conduction band (CB) state and the highest states in the heavy-hole (HH) and the light-hole (LH) band, respectively. By applying laser pulses with frequencies tuned either to the HH or the LH exciton energy the corresponding excitons can be created, destroyed, or manipulated. Initially the QD is in its ground state while the Mn spin is set to its lowest value $M_z = -\frac{5}{2}$, which could be prepared by adding a magnetic field at low temperatures. Because of the long relaxation time of the Mn spin in the range of tens of nanoseconds [5], in the absence of excitons this initial state is conserved on the time scales relevant for our studies.

Figure 1 shows a switching sequence for the Mn spin which, by excitation with a series of 100 fs laser pulses on the HH and LH transitions, is driven into each of the six eigenstates of M_z. In the lower part the pulse amplitudes on the HH and LH transitions are plotted while the upper part shows the occupations of the six Mn spin states. The solid lines refer to a calculation where a finite radiative lifetime of the bright excitons with a typical value of 250 ps [5] has been included, for the dashed lines recombination processes have been neglected. The shaded areas indicate switching times while in the white time intervals no laser pulses are applied. As seen from the occupations in the white intervals, starting from $M_z = -\frac{5}{2}$ the Mn spin indeed reaches all other states. These white intervals between the shaded switching times have mainly been inserted to demonstrate that here the Mn spin indeed remains in each of the states. When they are removed the total switching sequence takes about 100 ps. Although this is of the same order of magnitude as the radiative lifetime, it turns out that the exciton decay only marginally deteriorates the switching efficiency.

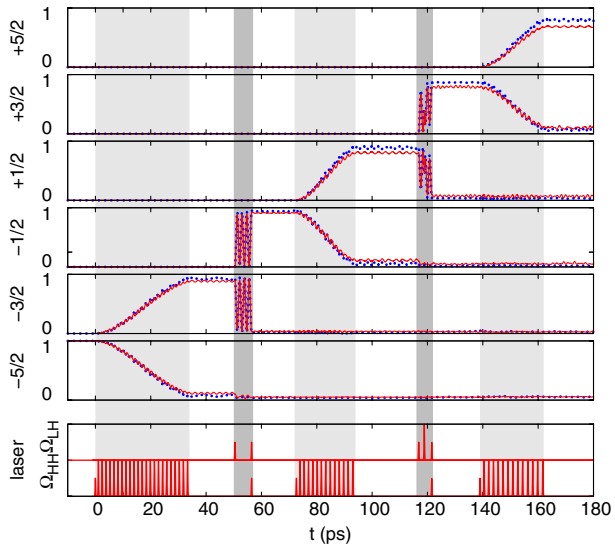

FIG. 1 (color online). Occupation of the six different states of the Mn spin and time sequence of the laser pulses on the LH and HH exciton transitions with (solid) and without (dashed) recombination. The shaded areas denote the switching processes while in the white areas no laser pulses are applied.

A clear drawback is the large number of pulses, especially on the HH exciton transition. However, as will be explained below, the spin dynamics strongly depend on magnetic fields. A switching sequence for a field of 9 T in Faraday geometry is shown in Fig. 2. Now a single pulse on the HH transition already initiates a complete flip to the next Mn spin state, and for the other switching processes only few pulses are necessary. Also the total switching time decreases to about 35 ps. On this time scale there is essentially no difference between the curves with and without radiative decay.

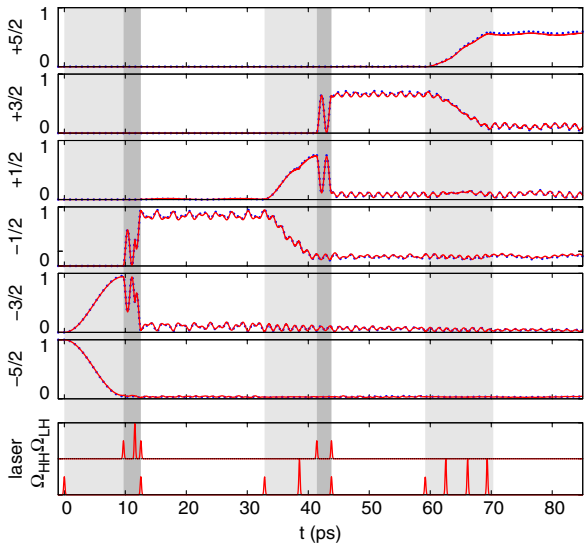

FIG. 2 (color online). Same as Fig. 1 but in the presence of a magnetic field of 9 T in Faraday geometry.

How can these switching dynamics be understood? For this purpose let us first describe in some more detail the QD model. Following Ref. [15], the CdTe QD is modeled by a square well potential with the dimensions $L_x = L_y =$ 7 nm and $L_z = 2$ nm. The CB states are s-like with a total angular momentum of $s = \frac{1}{2}$ and thus $s_z = \pm\frac{1}{2}$, while the p-like valence band (VB) states have a total angular momentum of $j = \frac{3}{2}$. In our case of a flat dot with $L_z \ll L_x$, L_y and $L_x = L_y$ the highest valence band state is the HH state with $j_z = \pm\frac{3}{2}$ and, as long as no strongly anisotropic strain is present [10], VB mixing is negligible here [16]. The LH state with $j_z = \pm\frac{1}{2}$ is below the HH ground state. For simplicity we neglect VB mixing also here, which requires a sufficiently large energy separation of the LH state from those excited HH states to which it is coupled, e.g., by the $\mathbf{k}\cdot\mathbf{p}$-Hamiltonian. From these electron and hole states excitons are formed, which are coupled to the light field in the usual dipole and rotating wave approximation [17]. Because of the selection rules the four HH exciton states consist of two bright ($H \pm 1$) and two dark states ($H \pm 2$). Analogously, there are two bright ($L \pm 1$) and two dark ($L \pm 0$) LH excitons. (We denote the exciton states by the valence band label (H, L) and the angular momentum $j_z + s_z$. In the case of the dark LH excitons with $j_z + s_z = 0$ the sign refers to the sign of j_z.) Including the ground state (0) and all states with two excitons, namely, the heavy-hole (HH0) and the light-hole (LL0) biexciton and four mixed HH-LH biexcitons (HL $\pm$ 2, HL $\pm$ 1), the total number of QD states adds up to 15.

This QD is doped with a single Mn atom with total angular momentum $M = \frac{5}{2}$ and thus six spin orientations M_z. It interacts with electrons and holes via the standard exchange coupling $\mathcal{H}_{c\text{-Mn}} = J_e\mathbf{M}\cdot\mathbf{S}_e(\mathbf{r}) + J_h\mathbf{M}\cdot\mathbf{S}_h(\mathbf{r})$, where $\mathbf{S}_{e/h}(\mathbf{r})$ denote the carrier angular momentum densities at the location of the Mn atom and $\mathbf{M}$ is the Mn spin. This interaction both lifts the degeneracy between bright and dark exciton states and induces correlated spin flips, where the spin of a carrier increases by one while the Mn spin decreases by one or vice versa. The electron-hole (e-h) exchange interaction $\mathcal{H}_{e\text{-}h} = J_{e\text{-}h}\mathbf{S}_e\cdot\mathbf{S}_h$ gives rise to a further splitting between bright and dark exciton states and can also induce spin flips in the exciton system [18]. The complete basis of the system is formed by the 90 states $|X; M_z\rangle$, where X refers to any of the 15 exciton states and M_z denotes the Mn spin state. The dynamical evolution is given by the Liouville–von Neumann equation for the density matrix elements $\rho_{n,n'}$, where n refers to any of the states $|X; M_z\rangle$. To include the finite radiative lifetime of bright excitons relaxation terms for the corresponding occupations (diagonal elements) and dephasing terms for the coherences (off-diagonal elements) have been added in the standard way [19]. The resulting equations of motion have been solved numerically.

To clearly understand the physics behind the switching protocol presented above let us now analyze separately the

different switching steps. First we will concentrate on the HH exciton subsystem coupled to the Mn spin. The e-h exchange interaction, the LH states and also the radiative decay are neglected in this part. In a pure HH system the hole spin, being $j_z = \pm\frac{3}{2}$, is pinned. When the QD is excited by a σ^--polarized laser pulse with a pulse area of π, the $(H-1)$-exciton is created. The resulting state $|1\rangle = |H-1; -\frac{5}{2}\rangle$ is coupled via the e-Mn exchange interaction to the state $|2\rangle = |H-2; -\frac{3}{2}\rangle$. In Fig. 3(a) we have plotted the occupations of the exciton states 0, $H-1$, and $H-2$ as well as the expectation value of the Mn spin for a single pulse excitation as a function of time. After the pulse there is no more coupling to the ground state; the system is effectively reduced to a two-level system consisting of bright and dark exciton states. The exchange interaction induces Rabi-type oscillations which are off-resonant because of the energy difference ΔE between these states caused by the Ising-type part of the exchange interaction. According to the theory of two-level systems, the occupation P_{H-2} of the dark exciton state thus should follow $P_{H-2} = (\xi_e/\hbar\Omega)^2 \sin^2(\Omega t/2)$ with $\xi_e = 2\langle H-1; -\frac{5}{2}|H_{c\text{-Mn}}|H-2; -\frac{3}{2}\rangle$ and the Rabi frequency $\hbar\Omega = \sqrt{(\Delta E)^2 + |\xi_e|^2}$, which is indeed in agreement with the result shown in Fig. 3(a).

Because of the off-resonant nature of this Rabi oscillation no complete spin flip is achieved. However, it turns out that the Mn spin can be efficiently controlled by exciting the QD with a pulse train. After the first π pulse creating a $H-1$ exciton a series of 2π pulses is applied at time intervals given by half the Rabi period. The resulting occupation of the $H-1$ exciton state and the expectation value of the Mn spin $\langle M_z\rangle$ are shown in Fig. 3(b). The π pulse creates the exciton and the occupation starts to oscillate. When the first minimum is reached, a 2π pulse is applied. During this pulse the exciton occupation drops to zero and then returns to its value before the pulse. However, the pulse creates a phase jump of π in the coherence $\langle H-2; -\frac{3}{2}|\rho|H-1; -\frac{5}{2}\rangle$ between bright and dark state and, as a result, the upper and lower bounds of the Rabi oscillation are shifted. The Mn spin follows this change and now oscillates between higher values. Without further pulses this oscillation continues, as shown by the thin solid line. Another 2π pulse again shifts the bounds of the oscillation and further reduces the occupation of the $H-1$ state. After five pulses the bright exciton has nearly completely turned into a dark exciton and the Mn spin has almost reached the value of $-\frac{3}{2}$.

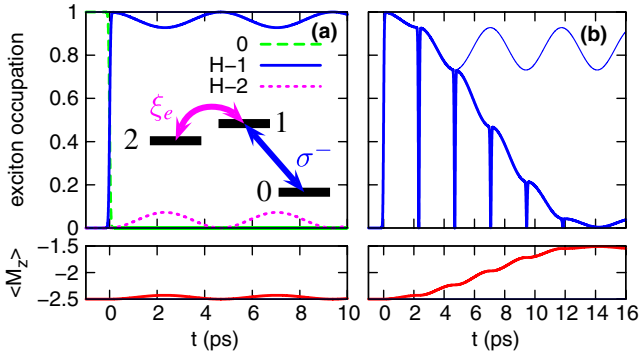

FIG. 3 (color online). (a) Occupation of the exciton ground state 0, the bright $(H-1)$ and dark $(H-2)$ HH exciton states (upper panel) and expectation value of the Mn spin $\langle M_z\rangle$ (lower panel) for an excitation by a single σ^--polarized π-pulse; (b) Occupation of the bright exciton state $H-1$ (upper part) and expectation value of the Mn spin $\langle M_z\rangle$ (lower part) for excitation by a train of laser pulses. Thin line: occupation of the exciton state after excitation with only two pulses.

When comparing Fig. 3(b) with the first switching sequence of Fig. 1 we notice that there a much larger number of pulses was necessary to flip the Mn spin from $M_z = -\frac{5}{2}$ to $M_z = -\frac{3}{2}$. This difference is mainly caused by the e-h exchange interaction, which in the full calculation has been included. It gives rise to an additional splitting between bright and dark exciton and thus to even more off-resonant Rabi oscillations which results in a higher Rabi frequency and a smaller amplitude of the oscillations. However, by a magnetic field applied in the z direction the energy splitting ΔE can be varied. In our case this splitting vanishes for a magnetic field of 9 T. Then the Rabi oscillation is resonant and a single pulse is sufficient to drive the exciton completely into the dark state, as is shown in Fig. 2. The spectral analogue of these Rabi oscillations, an anticrossing between bright and dark excitons, has indeed been seen in the magnetoluminescence of such QDs [7].

In the HH exciton system, due to the pinning of the HH spin, a further increase of $\langle M_z\rangle$ is not possible. However, by including LH excitons the spin flip dynamics can be continued since here both electron and hole spin can flip. If the LH exciton is created in addition to a HH exciton already present in the QD, a mixed biexciton is formed in which the CB state is completely filled. Thus, only the LH spin can flip. This is shown in Fig. 4(a), where we have plotted the occupations of the biexciton states HL $-$ 2 and HL $-$ 1 together with the expectation value of the Mn spin for a system which is initially in the dark state $|H-2, -\frac{3}{2}\rangle$ and then excited by a σ^+-polarized π-pulse tuned to the LH exciton energy. We again observe exchange-induced off-resonant Rabi oscillations, but with a higher amplitude and larger frequency than in the HH case. This is both due to the stronger exchange coupling of the holes and the smaller energy splitting between the two states.

We can now combine the dynamics in the HH and the LH exciton system to achieve multiple spin flips of the Mn atom. The path which is taken for this purpose is sketched in the reduced energy level scheme shown in Fig. 4(b). We start from the ground state $|0, -\frac{5}{2}\rangle$. By first creating a $(H-1)$ exciton and then applying a suitable sequence of 2π-pulses tuned to the HH transition we drive the system into the dark exciton state $|H-2, -\frac{3}{2}\rangle$. Now we apply a

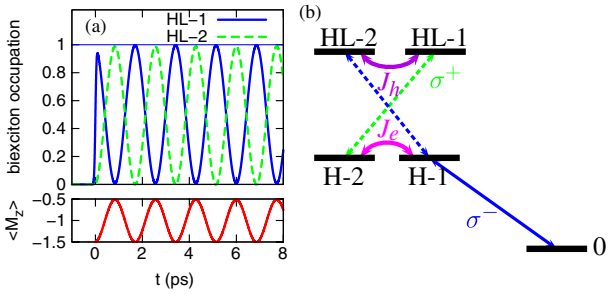

FIG. 4 (color online). (a) Occupation of the mixed biexciton states HL − 2 and HL − 1 (upper part) and expectation value of the Mn spin $\langle M_z \rangle$ (lower part) for the case of excitation of a system prepared in the state $|H-2,-\frac{3}{2}\rangle$ by a σ^+-polarized π pulses at $t = 0$ tuned to the LH transition. (b) Energy scheme of the exciton states and the couplings by laser pulses ($\sigma^{\pm}$) and carrier-Mn exchange interaction ($J_{e/h}$) for the switching process discussed in the text.

σ^+-polarized π-pulse tuned to the LH transition creating a mixed biexciton in the state $|\mathrm{HL}-1,-\frac{3}{2}\rangle$. By means of the h-Mn exchange interaction this state is coupled to the state $|\mathrm{HL}-2,-\frac{1}{2}\rangle$ and thus Rabi oscillations are initiated. If necessary we again apply a sequence of σ^+-polarized 2π-pulses to drive the system almost completely into the state $|\mathrm{HL}-2,-\frac{1}{2}\rangle$. This biexciton now consists of two optically active excitons, which can be removed by two σ^--polarized π-pulses, one tuned to the LH and one to the HH transition, and we end up in the state $|0,-\frac{1}{2}\rangle$. As a result of the whole process the Mn spin has been increased by two. We have created excitons by a σ^-- and a σ^+-pulse while we have removed them by two σ^--pulses. Thus, effectively the angular momentum has been transferred from the light to the Mn spin. Since the exciton system has returned to its ground state the whole process can be repeated to further increase the Mn spin.

The success of the switching seen in Figs. 1 and 2 is based on this protocol: (i) creation and manipulation of a HH exciton increases the Mn spin by one; (ii) creation and manipulation of an additional LH exciton again increases the Mn spin by one; (iii) removal of both excitons returns the QD to the ground state. Then the process can start again, such that all Mn spin states can be reached. Since the whole process occurs on times shorter than typical radiative lifetimes the effect of this process is weak (Fig. 1) or almost invisible (Fig. 2). Spin relaxation times are even longer [5,20]. LH excitons might be more sensitive to relaxation processes because the LHs can relax to HHs, but they are present in the QD only for a few ps before they are removed again. Thus, also here only a limited influence of relaxation processes is expected. To achieve a physically transparent and easily interpretable protocol we have restricted ourselves to sequences consisting only of π and 2π pulses and to a QD where the holes have a well-defined angular momentum. Optimizations by allowing for more general pulse shaping may be possible and refinements will be necessary if in a specific QD the hole states exhibit a strong VB mixing, but these generalizations are beyond the scope of this Letter.

In summary, we have shown that the spin state of a single Mn atom in a QD can be selectively controlled by manipulating the exciton states with ultrafast laser pulses. By using pulse sequences both on the HH and the LH exciton transitions all six possible spin states can be reached. The switching process can be optimized by applying a magnetic field which brings the Rabi oscillation in or close to resonance. We thus have presented a technique for an all-optical switching of the spin state of magnetic atom in a QD on a picosecond time scale.

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