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The role of magnetic fields in Faraday and Voigt geometry

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Dynamics of a single Mn spin in a quantum dot: The role of magnetic fields in Faraday and Voigt geometry

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Abstract. A theoretical analysis of the optically driven spin dynamics of a single Mn atom in a quantum dot in the presence of magnetic fields in different geometries is presented. When a magnetic field in Faraday configuration is applied, the Mn spin can be flipped from its initial state into each of its six spin eigenstates via optical excitation and manipulation of excitons. In the case of a magnetic field in Voigt configuration, the presence of an exciton leads to a precession of the Mn spin around the combination of external and exchange-induced magnetic field. By using optical pulses this precession can be switched on and off.

1. Introduction
Spin systems are attractive candidates for the implementation of a quantum logic. While usually quantum gates are built from two-level systems, i.e., qubits, it has recently been shown that it might be advantageous to employ higher-dimensional Hilbert spaces [1]. In a solid state environment such a higher-dimensional Hilbert space can be realized by a Mn atom which has a spin of \( J = \frac{5}{2} \) and thus six possible orientations \( M_z \). Quantum dot (QD) structures doped with a single Mn atom have recently been fabricated both in II-VI [2] and III-V materials [3]. In both cases, due to the strong exchange interaction between exciton and Mn spin, the exciton line in the photoluminescence (PL) spectra splits into a series of lines. Besides splitting the exciton line, the exchange interaction also gives rise to simultaneous spin flips of electron or hole and Mn. Thus, even if there is no direct coupling of the Mn spin to the light field, an indirect manipulation of the Mn spin state becomes possible via the optical excitation of excitons. This has recently been demonstrated experimentally in single [4] and in double QDs [5]. There the spin transfer was based on incoherent relaxation or transfer processes resulting in typical time scales of the order of several nanoseconds. In contrast, our scheme is based on the coherent light-induced dynamics of the exciton-Mn system resulting in switching times in the picosecond regime [6]. Here we will discuss the role of a magnetic field applied either in Faraday or in Voigt configuration on the Mn spin dynamics.

2. Model system
We consider a single CdTe quantum dot doped with a single Mn atom. For the electronic system we take into account the lowest conduction band state (\( S^c_z = \pm 1/2 \)) as well as the highest state...
in the heavy hole (HH, \(S^h_z = \pm 3/2\)) and in the light hole (LH, \(S^h_z = \pm 1/2\)) band, respectively. All these electron and hole states are two-fold degenerate. The electronic system thus consists of the ground state, four HH and four LH single exciton states, a HH and a LH biexciton state as well as four mixed HH-LH biexciton states, adding up to 15 states. The coupling of the excitons to the light field is treated in the usual rotating wave and dipole approximation. We assume Gaussian laser pulses with a full width at half maximum of 100 fs; a circularly polarized \(\pi\) pulse creates a single spin-polarized exciton in the system. The laser pulse frequency is tuned either to the HH exciton or the LH exciton transition. A HH-LH splitting of 40 meV has been assumed such that both transitions can be selectively addressed. The magnetic field \(\vec{B}\) couples via the Zeeman terms \(H_Z = \mu_B \left( g_{Mn} \vec{M} + g_e \vec{S}^e + g_h \vec{S}^h \right) \cdot \vec{B} \) with the g-factors \(g_{Mn}, g_e, \) and \(g_h\) and \(\mu_B\) denoting the Bohr magneton. The exchange interaction is of the usual Heisenberg type \(H_{exc} = j_e \vec{M} \cdot \vec{S}^e + j_h \vec{M} \cdot \vec{S}^h + j_{eh} \vec{S}^e \cdot \vec{S}^h\), consisting of electron-Mn, hole-Mn, and electron-hole contribution with the respective coupling constants \(j_e, j_h, \) and \(j_{eh}\). With the identity \(\vec{M} \cdot \vec{S}^e = M_z S^e_z + \frac{1}{2}(M_+ S^e_+ + M_- S^e_-)\) (and analogous expressions for the other contributions) we clearly see the two different types of terms: The Ising type term leading to the energy splitting and the terms leading to correlated spin flips of electron and Mn spin. PL spectra for both Faraday and Voigt configurations have been measured and interpreted in Ref. [7]. A model based on these ingredients has been successfully used to model the observed PL spectra [8]. With this Hamiltonian the Liouville-von Neumann equation for the density matrix of the coupled exciton-Mn system has been set up and solved numerically.

3. Spin flip in Faraday configuration

Let us first consider the case of a magnetic field of \(B = 6\) T in Faraday configuration. Since the magnetic field lifts the degeneracy of the Mn spin states, the ground state is now given by \(M_z = -5/2\). Starting from this initial state we apply a series of laser pulses, first on the HH exciton and then on the LH exciton transition, as plotted in the lower part of Fig. 1. The upper part shows the occupations of the three lowest Mn spin states \(M_z = -5/2, -3/2, \) and \(-1/2\). With the first \(\sigma^-\) circularly polarized pulse (pulse area \(\pi\)) a spin polarized \(H - 1\) exciton is created at time \(t = 0\). We denote the exciton states by their valence band type and their angular momentum \(S^h_z + S^e_z\). In a pure HH exciton the hole spin, being \(S^h_z = -3/2\), is pinned. The electron, however, which is created in the \(S^e_z = +1/2\) state can undergo a spin flip by simultaneously flipping the Mn spin from \(-5/2\) to \(-3/2\). These spin flips manifest themselves as exchange-induced Rabi oscillations between the combined exciton-Mn states \(|H - 1, M_z = -5/2\rangle\).
Figure 2. Expectation value of the Mn spin in the three directions $\langle M_x \rangle$, $\langle M_y \rangle$ and $\langle M_z \rangle$ as well the occupation of the exciton state as a function of time for the case of a magnetic field of 6 T applied in Voigt configuration. (a) Excitation with $\sigma^+$ pulses; (b) with $\sigma^-$ pulses.

and $|H-2, M_z = -3/2\rangle$, which are off-resonant due to the energy difference between the states and therefore do not lead to a complete flip. With a series of $2\pi$-pulses, however, we can push the system almost completely into the state $|H-2, M_z = -3/2\rangle$, since each $2\pi$ pulse creates a phase jump in the coherence between the involved states. Thus we have effectively switched the Mn spin from $M_z = -5/2$ to $M_z = -3/2$. Indeed, in Fig. 1 we observe that after 7 pulses the occupation of $-5/2$ is nearly zero while the occupation of $-3/2$ has raised to nearly one. Now the exciton is in the dark state $H-2$, which is not accessible anymore by laser pulses resonant to the HH exciton transition. To overcome this bottleneck we apply a $\sigma^+$ polarized $\pi$-pulse on the LH transition and create a combined biexciton $HL-1$. Now the conduction band state is filled with two electrons, the HH spin is still pinned, but the LH can perform a spin flip accompanied by a simultaneous Mn spin flip to switch the Mn spin from $-3/2$ to $-1/2$. Because the coupling of the Mn spin to the holes is four times stronger than to the electrons and the energy splitting between the involved states is much smaller, the exchange-induced Rabi oscillation is much faster and stronger than was the case for the electron spin flip. For this oscillation, as can be seen in Fig. 1, only one $2\pi$ pulse is needed to push the Mn spin state almost completely to $-1/2$. The biexciton now consists of two bright excitons, so two $\pi$ pulses, one resonant to the HH and the other to the LH transition, are applied to bring the electronic system back to its ground state. This scheme can be repeated to switch the Mn spin into all of its spin eigenstates [6]. While this scheme works for any magnetic field strength, the details (number of pulses, total switching time, etc.) strongly depend on the field [9].

4. Spin dynamics in Voigt configuration
The situation is very different when a magnetic field in Voigt configuration is applied. We again take a field of $B = 6$ T, but now oriented in $x$-direction. Without exciton the magnetic field defines the quantization axis for the Mn spin, which therefore in the ground state has the spin $M_x = -5/2$. With a single $\sigma^+$ polarized $\pi$-pulse at time $t = 0$ we now create a $H+1$ exciton in the system. In Fig. 2(a) the expectation values of the three components of the Mn spin as well as the occupation $\rho_X$ of the bright exciton states are shown. We start with $\langle M_x \rangle = -5/2$ and
\( \langle M_y \rangle = \langle M_z \rangle = 0 \). After the excitation all three expectation values of \( \vec{M} \) start to oscillate with a period of \( T = 4.2 \) ps. The reason for this behavior is that the exchange interaction effectively gives rise to an additional magnetic field. The total effective field for the Mn spin is then given by

\[
\vec{B}_{\text{eff}}^{\text{Mn}} = \vec{B} + \frac{j_e}{g_{\text{Mn}}\mu_B} \vec{S}^e + \frac{j_h}{g_{\text{Mn}}\mu_B} \vec{S}^h .
\] (1)

Like in the previous section a HH with spin \( S^h_z = \pm 3/2 \) cannot flip. Its quantization axis is given by the growth direction, therefore the hole always gives a contribution to the effective field which points in z-direction. The electron contribution does not have a fixed direction because the electron spin precesses as well. However, as can be seen in the bottom part of the Figure, the exciton essentially remains in the \( H + 1 \) state, thus also the electron contribution to the effective field is mainly in z-direction. Then the effective field is in the \( xz \)-plane. For our parameters it has a value of \( B_{\text{eff}}^{\text{Mn}} = 8.4 \) T and is oriented at an angle of 45° with respect to the \( x \)-axis. This estimate is completely in agreement with the Mn spin dynamics seen in Fig. 2(a): The \( x \)- and \( z \)-components of \( \langle \vec{M} \rangle \) oscillate between \(-5/2 \) and \( 0 \) while the \( y \)-component oscillates symmetrically around zero. Much like the Mn spin also the electron experiences an effective magnetic field. Here, the strongest contribution comes from the electron-hole exchange interaction. The oscillation of the Mn spin gives rise to an additional oscillating contribution in this effective field. The resulting precession of the electron is seen in the oscillations of the occupation of the bright exciton state [bottom part of Fig. 2(a)]. After three periods of the oscillation, i.e. at \( t = 12.6 \) ps, we apply a second \( \pi \) pulse to annihilate the exciton and thus switch off the precession. At this time the Mn spin has rotated back to its initial position. With the annihilation of the exciton, the exchange-induced magnetic field contribution also ends and only the external magnetic field acts on the Mn spin. Thus the Mn spin is now again approximately in an eigenstate and almost no further precession takes place. Figure 2(b) shows that exciting the system with a \( \sigma^- \) pulse leads to exactly the same type of dynamics, only \( \langle M_y \rangle \) and \( \langle M_z \rangle \) have the opposite sign.

5. Conclusions

We have analyzed the dynamics of a single Mn spin in a single QD under the influence of a magnetic field in either Faraday or Voigt configuration. In Faraday configuration by starting with an initial spin of \( M_z = -5/2 \) we were able to switch the Mn spin by 2 into the state \( M_z = -1/2 \), while the exciton system returned to its ground state. In Voigt configuration the initial Mn spin points perpendicular to the \( z \)-direction and so in the initial state \( \langle M_z \rangle = 0 \). If now an exciton is created in the system, \( \langle M_z \rangle \) starts to precess. By creation and annihilation of an exciton the effective magnetic field is changed and correspondingly the precession of the Mn spin can be switched on and off on the time scale of the optical pulse.

References