Fast preparation and detection of Mn spin states in a magnetically doped quantum dot

Doris E. Reiter\textsuperscript{1}, Vollrath Martin Axt\textsuperscript{2}, Tilmann Kuhn\textsuperscript{1}

\textsuperscript{1}Institut für Festkörpertheorie, Westfälische Wilhelms-Universität, 48149 Münster, Germany
\textsuperscript{2}Theoretische Physik III, Universität Bayreuth, 95440 Bayreuth

E-mail: Doris.Reiter@uni-muenster.de

Abstract. The spin dynamics of a single Mn spin in a single quantum dot under the excitation with ultra fast laser pulses is studied. We develop a switching protocol that is able to prepare stable superpositions of states with different Mn spin quantum numbers. Furthermore the optical signal in a pump-probe type setup is calculated which can be used to read out the Mn spin state.

1. Introduction

For applications in quantum information it is of crucial importance to be able to prepare both quantum states with well defined quantum numbers as well as stable coherent superposition states. An attractive system for corresponding applications is the spin of a single Mn atom in a II-VI quantum dot (QD) as has been fabricated recently \cite{1}, since the Mn spin showed very long decoherence times in optical experiments \cite{2}. The optical manipulation of the Mn spin so far was based on incoherent relaxation or transfer mechanisms on the timescale of nanoseconds \cite{3, 4}. We have recently proposed a protocol based on coherent manipulations by which spin eigenstates of the the Mn atom could be prepared on a picosecond time scale. In this paper we present an extension of this protocol to create coherent superpositions of these eigenstates.

2. Theoretical background

We model a single CdTe QD doped with a single Mn atom following the approach in Ref. \cite{5}. We take into account one conduction band (CB) and two valence bands (VBs). Electrons in the CB have a total angular momentum of $S^e = \frac{1}{2}$ with $S^e_z = \pm \frac{1}{2}$, while the total angular momentum of the holes is $S^h = \frac{3}{2}$ splitting into the heavy hole (HH) states with $S^h_z = \pm \frac{3}{2}$ and the light hole (LH) states with $S^h_z = \pm \frac{1}{2}$. The HH-LH splitting is taken as 40 meV. Together the excitonic system comprises of 15 states including the ground state with no exciton $|0\rangle$, the single exciton states consisting of one HH and one electron $|H \pm 2\rangle$ and $|H \pm 1\rangle$, the single exciton states with one LH and one electron $|L \pm 1\rangle$ and $|L \pm 0\rangle$ (the sign in the latter refers to the LH spin), the biexciton states formed by either two HH or LH excitons $|HH0\rangle$ and $|LL0\rangle$, as well as four combined biexciton states composed of one HH exciton and one LH exciton $|HL \pm 2\rangle$ and $|HL \pm 1\rangle$. All states are labeled by their hole state and their total angular momentum. The Mn has six spin states with $M_z = \pm \frac{5}{2}, \pm \frac{3}{2}, \pm \frac{1}{2}$. The 90 states of the full system are then labeled $|X; M_z\rangle$ with the exciton state $|X\rangle$ and the Mn spin $M_z$. The electron and hole spin couple
to the Mn spin via the exchange interaction $H_{\text{exc}} = j_e \vec{M} \cdot \vec{S}^e + j_h \vec{M} \cdot \vec{S}^h$. For the calculations $j_h = 0.423$ meV and $j_e = -j_h/4$ are taken. The exchange interaction can be separated into two types of terms: using type terms $\sim j_e/h M_z S^e_z/h$, which lead to an energy shift of the states, and spin flip terms $\sim j_e/h \frac{1}{2}(M_z S^e_z/h + M_z S^e_{-z}/h)$. Due to the latter term simultaneous flips of the Mn spin and the electron/LH spin can occur. Note that a spin flip involving HHs is not possible by this term. Light coupling to the exciton system is treated in the standard dipole and rotating wave approximation. We consider Gaussian pulses with a full width at half maximum (FWHM) of 100 fs and a $\pi$ pulse is set to create one exciton. The single exciton states can be classified into bright (optically active) with angular momentum $\pm 1$ and dark (optically not active) states. Additionally the $e - h$ exchange interaction as well as Zeeman coupling to a magnetic field are taken into account. Optical signals to simulate a pump-probe setup, where a sequence of pump pulses excites the system, which is then probed by a weaker probe pulse with a FWHM of 10 ps, are calculated in the standard way. The spectrum is obtained by a Fourier transform with a damping of 20 ps.

3. Results

The absorption spectrum of a quantum dot containing a single Mn atom has a very characteristic feature: The exciton line of the lowest excitation splits into a set of six lines even at zero magnetic field according to the six orientations of the Mn spin [1]. A calculated absorption spectrum is shown in Fig. 1 for a magnetic field in Faraday configuration of $B_z = 6$ T and $\sigma^-$ polarized light resonant of the HH exciton transition. The six peak structure is clearly observable. Each of the six peaks can be identified with the Mn spin state $M_z = \pm \frac{5}{2}$ to $M_z = -\frac{5}{2}$ from left to right [5]. Some small side peaks appear due to the coupling of electron and Mn spin.

In the time domain the spin flip terms manifest themselves as exchange induced Rabi oscillations between the initially excited state and the state, where electron and Mn spin have changed by one. Usually the two states involved are not in resonance, such that this oscillation has an amplitude smaller than one. However, by applying a series of $2\pi$ pulses a complete flip can be achieved [6]. Using this feature a switching scheme was developed to address selectively all states of the Mn spin [7]. In this paper a modified switching scheme to create superpositions between states with different Mn spin is proposed.

Starting from the initial state $|0; -\frac{5}{2}\rangle$ a $\sigma^-$ polarized $\pi$ pulse on the HH exciton transition excites the system to $|H - 1; -\frac{5}{2}\rangle$. Note that the Mn spin is not affected by the laser excitation. In this state the electron can perform a spin flip while increasing the Mn spin by one, ending up in the state $|H - 2; -\frac{3}{2}\rangle$. If the spin is only halfway flipped, the system is in a superposition between the states $|H - 1; -\frac{5}{2}\rangle$ and $|H - 2; -\frac{3}{2}\rangle$. Now a $\sigma^+$ polarized pulse on the LH exciton transition is applied. This does not act on state $|H - 1; -\frac{5}{2}\rangle$, but excites the dark state $|H - 2; -\frac{3}{2}\rangle$ into the combined biexciton state $|HL - 1; -\frac{3}{2}\rangle$. In the latter state the CB state is filled, thus electrons and HH cannot change the Mn spin. Only the LH can flip and increase the Mn spin state, changing the state to $|HL - 2; -\frac{3}{2}\rangle$. This flip is much faster than the electron flip, such that $|H - 1; -\frac{5}{2}\rangle$ does not change. The combined biexciton $|HL - 2\rangle$ is optically active and by two $\pi$ pulses with $\sigma^-$ polarization, one resonant on the HH transition and one resonant on the

![Figure 1. Calculated absorption spectrum at a magnetic field of $B_z = 6$ T for $\sigma^-$ polarized light. In the initial state the six Mn spin states are taken to be equally occupied.](image)
LH transition, it is de-excited to $|0; -\frac{5}{2}\rangle$. The $\pi$ pulses likewise act on the other state $|H-1; -\frac{5}{2}\rangle$, turning the HH into a LH exciton via the ground state. Thus after the two $\pi$ pulses the system is in the superposition of the states $|L-1; -\frac{5}{2}\rangle$ and $|0; -\frac{1}{2}\rangle$. Both states are stable, because in the first state the spins of all particles have their minimal value and thus cannot flip, while in the latter state no exciton is present. To switch the Mn spin further, the same combination of pulses can be used. While these pulses do not change the Mn spin state with $M_z = -\frac{5}{2}$, it increases the Mn spin $M_z = -\frac{1}{2}$ to $M_z = +\frac{3}{2}$. After the switching the exciton system goes back to the ground state for both states. Thus a coherent superposition between the states $|0; -\frac{5}{2}\rangle$ and $|0; +\frac{3}{2}\rangle$ has been established.

The creation of a superposition state can also be followed in a pump-probe setup. Figure 3 shows probe spectra for $\sigma^-$ polarized light on the HH exciton transition taken at different time delays $\tau$ during the switching sequence presented in Fig. 2(a). The time delay refers to the starting point of the pump pulse sequence. Thus negative time delays refer to the case when the probe pulse precedes the first pump pulse. Here no exciton is in the system and $M_z = -\frac{5}{2}$. Accordingly an absorption peak is observed in Fig. 2(a). The positions of the six peaks of the absorption spectrum in Fig. 1 are marked by thin vertical lines and indeed the single peak in the...
Figure 3. Probe spectra for different time delays $\tau$ as indicated for the switching sequence shown in Fig. 2(a).

Figure 4. Probe spectra after the switching sequence for differently weighted superpositions as shown in Fig. 2.

probe spectrum matches the rightmost line. In addition, well known spectral oscillations caused by the following pulses are observed. The second probe spectrum (b) at $\tau = 13$ ps is taken, when the first stable superposition between $|L-1; -\frac{5}{2}\rangle$ and $|0; -\frac{1}{2}\rangle$ has been created. In the spectrum two peaks occur, one at the third line from the right, which corresponds to $M_z = -\frac{1}{2}$, while the other does not match one of the six lines. Because a LH was added to the system, the probe pulse does not act on the ground state to single exciton transition, but on the transition between LH exciton and combined biexciton. Thus the line is shifted by exchange interactions. Again spectral oscillations are superimposed. The spectrum (c) is taken at $\tau = 37$ ps, when a superposition between $M_z = -\frac{5}{2}$ and $M_z = -\frac{1}{2}$ is created. Two absorption peaks are seen at the second line from the left and the rightmost line in agreement with the Mn spin states.

Figure 4 finally shows probe spectra at a delay after the switching has taken place for the three cases with different weights of the Mn spin states. In each spectrum two peaks are seen corresponding to the respective Mn spin states. The strength of the peaks reflects the occupation of states: in (a) both peaks are equally high, in (b) the right peak, corresponding to $M_z = -\frac{5}{2}$ is much smaller than the left peak corresponding to $M_z = +\frac{3}{2}$, and in (c) it is vice versa.

4. Conclusions
We have proposed a switching protocol based on coherent manipulations of a single Mn spin in a QD to create coherent superpositions of Mn spin states. The creation of a superposition could be monitored in probe spectra, where two peaks appear, their strength reflecting the weights of the states of the superposition.

References
[1] Besombes L et al. 2004 Phys. Rev. Lett. 93 207403
[2] Besombes L et al. 2008 Phys. Rev. B 78 125324
[3] Le Gall C et al. 2009 Phys. Rev. Lett. 102 127402
[4] Goryca M et al. 2009 Phys. Rev. Lett. 103 087401
[5] Fernández-Rossier J 2006 Phys. Rev. B 73 045301
[6] Reiter D E, Kuhn T and Axt V M 2009 Phys. Status Solidi B 246 779
[7] Reiter D E, Kuhn T and Axt V M 2009 Phys. Rev. Lett. 102 177403