Optical Manipulation of a Single Mn Spin in a CdTe-Based Quantum Dot

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Two coupled CdTe quantum dots, selected from a self-assembled system, one of them containing a single Mn ion, were studied by continuous wave and modulated photoluminescence, photoluminescence excitation, and photon correlation experiments. Optical writing of information on the spin state of the Mn ion has been demonstrated, using the orientation of the Mn spin by spin-polarized carriers transferred from the neighboring quantum dot. Mn spin orientation time values from 20 to 100 ns were measured, depending on the excitation power. Storage time of the information on the Mn spin was found to be enhanced by application of a static magnetic field of 1 T, reaching hundreds of microseconds in the dark. Simple rate equation models were found to describe correctly the static and dynamical properties of the system.

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One of the important research directions that may influence the future of information processing, especially of spintronics [1], is focused on physical phenomena occurring in nanoscale-size quantum objects. One of these objects, close to the ultimate limit for information storage miniaturization, is a single Mn atom in a semiconductor quantum dot (QD) [2,3]. After intensive studies of semimagnetic QDs containing many magnetic ions [4–8], single Mn atoms in CdTe [9] and InAs [10] QDs have been observed in photoluminescence (PL) experiments. Many experiments supplied substantial knowledge concerning the physical properties of single Mn atoms, especially in CdTe QDs. In particular, they revealed the strong influence of the position of the Mn atom in the QD, reflecting the symmetry of the system [11]. They also demonstrated an efficient optical readout of the Mn spin state [9]. Furthermore, the dynamics of this state has been studied in photon correlation experiments [12], revealing an important influence of photocreated carriers on the Mn spin relaxation. The writing and storing of the information on the Mn spin state has received less attention so far. These issues represent the focus of this work.

In particular, we demonstrate optical writing of information on the spin state of a single Mn ion and we test the stability of this state in the time range up to 0.2 ms.

CdTe QDs containing single Mn ions were grown by molecular beam epitaxy. A single layer of self-assembled QDs was deposited in a ZnTe matrix. Manganese was added by briefly opening the Mn effusion cell during the growth of the CdTe layer [13]. The opening time and the Mn flux were adjusted to achieve a large probability for the growth of QDs with a single Mn ion in each dot. The selection of single QDs was achieved by the spatial limitation of the PL excitation and detection to an area smaller than 0.5 μm in diameter, using a microscope objective immersed in pumped liquid helium. Continuous wave excitation was used either above the ZnTe barrier gap (at 457 nm) or by a tunable dye laser in the range 570–600 nm. Well-separated photoluminescence lines from individual QDs were observed in the low energy part of the PL spectrum. We were able to select numerous lines showing a PL pattern characteristic for the presence of a single Mn ion in a dot [2,9]. An example of such a spectrum is presented in Fig. 1(a). It contains six lines related to neutral exciton (X), equally spaced over a range of 1.5 meV. The splitting into six components is due to the exciton-Mn exchange interaction. At an energy about 11 meV below, the biexciton (XX) PL from the same QD is also split in 6 lines with the same value for the energetic splitting. As a

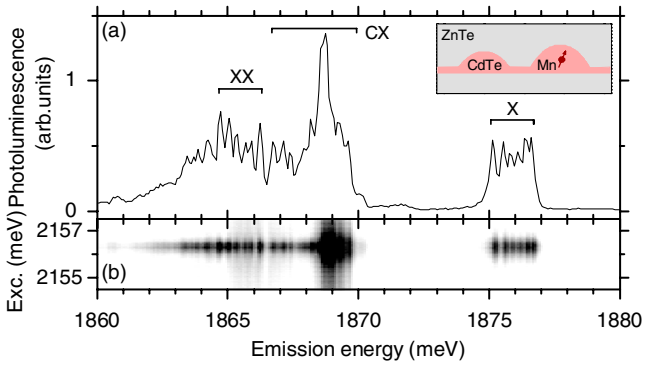

FIG. 1 (color online). (a) Photoluminescence spectrum of a single QD with a single Mn ion, excited by linear polarization with indicated features related to neutral exciton (X), biexciton (XX), and charged excitons (CX). (b) Photoluminescence excitation map (density plot of PL intensity versus excitation/emission photon energy) close to the resonance. Inset: Schematic diagram of the relative position of the absorbing (smaller) and the emitting (larger, containing Mn ion) QD.

test of the identification of these two transitions, photon correlation measurements were performed in a Hanbury Brown and Twiss setup [14] using two monochromators equipped with avalanche photodiode detectors. The obtained biexciton-exciton cross-correlation histograms (not shown) exhibit a bunching peak related to a radiative cascade, confirming the identification of both transitions and their common origin (the same QD). Between the two features there are series of lines related to charged excitons. We have performed systematic measurements of PL for various values of excitation power and photon energy. The results were coherent with the typical behavior for similar QDs [15]. Among the identified QDs with single Mn ions, we selected those exhibiting a sharp resonance in the photoluminescence excitation spectra, as shown in Fig. 1(b).

Similar resonances were found previously in CdTe/ZnTe QDs without manganese and interpreted as related to the transfer of excitons between a smaller and a larger QD [16]. They exhibit properties of the fundamental state of neutral exciton in CdTe QD: (i) they are very sharp, (ii) their energy is distributed over the range of the QDs band observed by macro-PL, (iii) they exhibit optical in-plane anisotropy (in zero magnetic field) as well as Zeeman splitting and diamagnetic shift (in high magnetic field) typical for neutral excitons observed in PL. The above properties exclude the wetting layer states, deep centers, and high excited states as possible explanations of the observed resonances. Additionally, we found that while the PL exciton lines exhibit characteristic sixfold splitting due to the interaction with single Mn ion, the absorption line is not split by this interaction [see Fig. 1(b)]. Moreover, all the observed charge states of the emitting QD share the same resonance energy. These arguments prove that the observed resonance is related to a different state than the state in the emitting QD. Therefore we conclude that the resonance is related to absorption in a QD with no manganese. The photocreated exciton is then transferred to a larger dot containing a single Mn ion. Note that finding such a pair of QDs is possible despite the quite low dot density (equal to about 5×10^9 cm^{-2}), because of a tendency to form groups of closely spaced dots, observed in our samples [17]. A more detailed discussion of the interdot transfer mechanism can be found in Ref. [16].

Experimentally, the transferred exciton conserves its spin (similarly as in Ref. [16]). It is clearly visible in Figs. 2(a)–2(d). The exciton PL spectra measured in the same circular polarizations of excitation and detection (copolarized) have higher intensity integrated over the 6 exciton lines than those measured in different polarizations (cross polarized). The lower intensity equals about 65% of the higher one for an excitation power of about 6 μW, which gives a lower bound for the polarization transfer efficiency.

The transfer efficiency depends on the details of the parameters of the coupled QDs (such as in-plane anisotropy and excitation transfer rate). In our case it is almost

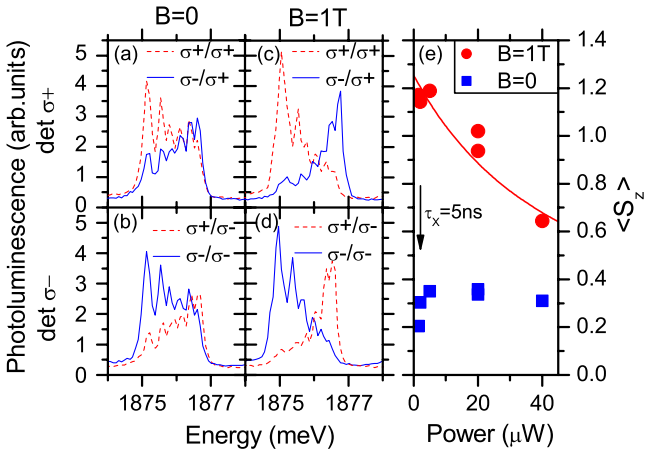

FIG. 2 (color online). (a)–(d) Photoluminescence spectra excited at the resonance, using indicated excitation-detection polarizations. Measurements taken at $T = 1.5$ K, and $B = 0$ T (a), (b) or $B = 1$ T (c),(d). (e) The mean spin of the Mn ion as a function of excitation power for $B = 0$ and $B = 1$ T compared with model described in text (solid line). The arrow indicates the power at which averaged time delay between capture of two excitons (τ_X) is equal to 5 ns.

independent of magnetic field, and decreases for higher excitation power due to the increased probability of biexciton formation. The presence of biexcitons leads to a suppression of the spin polarization transfer, since the biexciton singlet ground state cannot carry any spin memory.

The exciton polarization transfer is used as a tool for optical writing of information on the Mn spin. Carriers created by a defined circular polarization of light act on the Mn ion via the exchange interaction and orient its spin. The Mn spin orientation appears in Figs. 2(a)–2(d) as a nonuniform distribution of intensities between the 6 exciton lines, which reflects a nonequal probability of finding the Mn atom in each spin state. As the Mn spin polarization results from interaction with spin-polarized carriers, the spin polarizations of both species are related to each other. This relation will be discussed later. As a quantitative measure of the spin orientation we use the mean spin value of the Mn ion determined from the mean position of the PL sextet (weighted by intensity). An example variation of this value versus excitation power is shown in Fig. 2(e) ($B = 0$ data). It shows a maximum at about 0.1 mW/μm^2 and decreases for higher and lower power. The decrease at the high power corresponds to the increased probability of the biexciton formation and—as a consequence—a decrease of the efficiency of the Mn spin alignment. The decrease at low excitation power is due to the competition between the Mn spin relaxation and the spin orientation by the photocreated excitons. The relaxation becomes dominant when the average time delay between capture of two excitons is longer than the relaxation time. This condition provides an estimate of the relaxation time, being in our case of the order of a few nanoseconds. This value compares reason-

ably with the relaxation time determined for very diluted (Cd,Mn)Te quantum wells in the absence of magnetic field [18]. The fast relaxation was caused mainly by the hyperfine interaction with nuclear spin.

As shown in Ref. [18], the fast relaxation may be blocked by applying a small magnetic field, which suppresses the mixing of the Mn spin eigenstates by the hyperfine interaction. Therefore, in order to define pure spin states of the Mn ion, a magnetic field of about 1 T was applied. The field was weak enough to assure a negligible orientation of the Mn spin by its thermal distribution. This was checked when the QD was excited using linearly polarized light. The mean spin was then equal to about 0.2. In contrast, under circularly polarized excitation the mean spin was up to 1.2. The variation of the mean Mn spin versus excitation power at a fixed magnetic field and polarization of the light is shown in Fig. 2(e). Similarly to the zero-field case, it shows a decrease at high power, resulting from an increase of the probability of biexciton formation. However, no decrease is observed at low power, down to its lowest values. This may be seen as an argument for a long spin relaxation time.

The limit of low excitation power in a magnetic field corresponds to a particularly simple situation, when the only mechanism influencing the Mn spin state is the interaction with photocreated carriers present in the emitting dot. A simple optical orientation model can be tested in this situation. It is based on the assumption that any Mn state, characterized by its quantum number m, can be transferred to a state with m different by 1 with a fixed probability p_0 during the presence of a suitably polarized exciton in the QD. This gives a simple rate equation for the probability of the occupation of each Mn spin state: $dq_m/dt = p_0[q_{m-1}\varphi_{X_{-1}} + q_{m+1}\varphi_{X_{+1}} - q_m(\varphi_{X_{-1}} + \varphi_{X_{-1}})]$, where $\varphi_{X_{\pm 1}}$ are frequencies of arriving of excitons into the QD (with a suitable cutoff for $|m| = 5/2$). The transfer may be caused either by spin flip-flop with photocreated carriers or by a relaxation in the effective exchange field of the exciton. A detailed study of its mechanism and its exciton life-time-scale dynamics is beyond the scope of this Letter. Since the Mn reorientation is much slower than the exciton recombination (as will be shown further below), the two processes can be analyzed separately. Within the assumptions of the model, the steady state of Mn spin does not depend on absolute exciton transfer rate or p_0 value, but only on the relative numbers of spin-up and spin-down excitons arriving in the emitting dot.

To test the applicability of the model, we sum the up and down Mn spin transfer probabilities weighted by the distribution of Mn states read from the sextuplet spectrum of Figs. 2(c) and 2(d) and relative frequencies of arriving spin-up and spin-down excitons in the dot, known from the previously estimated exciton spin transfer efficiency. The up and down transfer rates thus obtained are equal with good accuracy, as required in a steady state. In other words, the exciton polarization transfer efficiency is directly correlated with the Mn spin state.

The model can be easily extended to take into account the influence of biexcitons, assuming that each of them contributes with equal probability $p_0/2$ to the Mn up and down spin flips. To determine the biexciton:exciton ratio of arrival rates in the QD, we use a rate equation model of exciton dynamics introduced by Regelman *et al.* [19]. We select a basis containing up to three excitons in the dot. Using the determined biexciton:exciton arrival rate ratio, the power dependence of Mn spin orientation was calculated, with the exciton spin transfer efficiency as a free parameter. An excellent agreement was obtained [Fig. 2(e)], with spin-up to spin-down probability ratio of 0.62. This value agrees well with the lower limit of the polarization transfer determined experimentally from the net polarization of the exciton line at low excitation power.

The spin relaxation time was analyzed in more detail in an experiment, in which the excitation and its polarization were modulated. This was achieved by passing the laser beam through acousto-optic and electro-optic modulators. Using the first one we were able to switch the excitation on and off. The second modulator allowed us to change the circular polarization of the laser. The switching time of both modulations was 10 ns. The modulators were driven by a set of generators synchronized with a time-resolved photon counting system (PicoQuant TimeHarp 200), which was used to record the temporal profiles of the PL signal. The excitation sequence was as follows: first the Mn spin was oriented using a σ^+ polarized excitation. Then the light was switched off for a controlled delay and subsequently the light with opposite circular polarization was switched on to accomplish the readout of the spin state. This pattern was repeated with a sufficiently low frequency to reach a steady Mn spin state each time the light was on. The Mn state was read by measuring the PL intensity of a selected component of the exciton sextuplet in a given circular polarization. Its evolution reflects the orientation of the Mn spin combined with the exciton optical orientation. The measurements were repeated for both circular polarizations of detection. An example temporal profile is presented in Fig. 3(a). It was obtained using σ^+ polarization for the detection and detecting at the energy of the lowest component of the exciton sextuplet.

Several features present in the profile may be used to determine different properties of the spin orientation. The first flat region of the profile is related to excitation with σ^+ polarization, the same as the detection polarization. When the excitation is switched off, the PL signal drops to zero and remains vanishing as long as the excitation is off. When subsequently a σ^- polarized excitation is turned on, the PL intensity first rises rapidly to an initial value determined by the Mn spin state and the efficiency of the exciton polarization transfer. If the Mn orientation is conserved, this value is given only by the exciton polarization transfer and should be equal to about 65% of the previous value for an excitation power of 6 μW [as marked in Fig. 3(a)]. Any loss of the Mn spin orientation would decrease this value. Figure 3(a) shows clearly that the Mn spin state immedi-

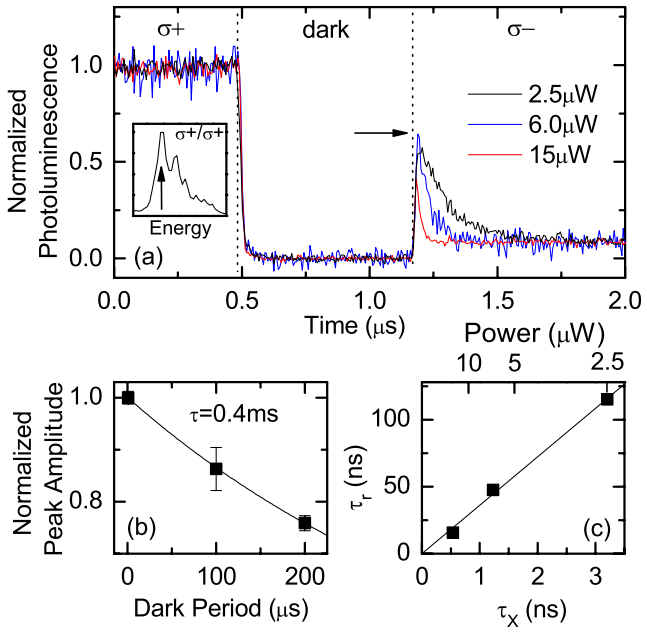

FIG. 3 (color online). (a) Temporal profile of the PL intensity at one of the six components of the exciton spectrum (indicated in inset) during the excitation sequence described in text for $B = 1$ T and indicated values of excitation power. The horizontal arrow indicates the ratio between PL intensity measured in copolarization and cross-polarization setup at excitation power equal to 6 μW. (b) PL intensity after turning on the excitation in σ^- polarization versus length of dark period. (c) The Mn reorientation time upon illumination in σ^- polarization (τ_r) versus averaged time delay between capture of two excitons (τ_X) and excitation power.

ately after the dark period is very close to that written by the light. The initial value of PL intensity is presented versus delay in Fig. 3(b). Its decrease allows us to roughly estimate the Mn spin relaxation time in the dark to be about 0.4 ms.

After the initial rapid increase, the PL intensity slowly decreases with a characteristic time of tens of nanoseconds. This decrease is related to the reorientation of the Mn spin by circularly polarized light. This process accelerates with increasing excitation power, as shown in Fig. 3(a). Its characteristic time is approximately inversely proportional to the excitation power. This property suggests that the simple model, introduced to describe the steady-state Mn spin orientation, can also be used to interpret its dynamics. In contrast to the steady-state case, now the rate at which excitons arrive in the emitting QD, as well as the spin-flip probability p_0, becomes important. We determine the former from the biexciton-exciton intensity ratio, using our rate equation model. The latter enters the model calculation as a free parameter. The result of such a calculation, presented in Fig. 3(c), describes well the experimental values, assuming $p_0 = 0.1$. It is also important to note that in our case the photocreated excitons are well defined and no free photocreated carriers are expected to contribute to the spin relaxation. We excite resonantly excitons in the neighboring QD with an excitation power sufficiently low to make any nonresonant processes negligible. Therefore, in contrast to Ref. [12], we do not consider the spin exchange with individual carriers surrounding the QD.

To conclude, we have demonstrated that the information can be written on the spin state of a single Mn ion in a QD using an orientation process, which exploits exciton spin transfer from a neighbor nonmagnetic QD. The orientation time varied between 20 and 100 ns over the used range of excitation power. We determined a storage time of information on the Mn spin. It is enhanced by application of a moderate static magnetic field and reaches hundreds of microseconds in the dark.

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