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Spin-Related Spectroscopy of CdTe-Based Quantum Dots

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This work contains a selection of our recent experimental results in the field of the spin-related spectroscopy of individual CdTe-based quantum dots. After a short description of the sample growth and experimental methods, optical measurements of the charge state dynamics are presented. Then the influence of in-plane anisotropy of the excitonic states of a quantum dot is discussed, followed by a description of experimental studies of information read-out and writing on quantum dot spin states. In particular, spin memory of a single Mn^{++} ion embedded in a CdTe quantum dot is quantitatively assessed. In an outlook part, perspectives opened by recently developed ZnTe lattice-matched Bragg reflectors are discussed.

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1. Introduction

Spin in semiconductor quantum dots (QDs) attracts attention as a medium for storage and manipulation of quantum information [1, 2]. Spectroscopic methods represent an efficient way to study the spin properties of the quantum dots. Cadmium telluride belongs to the family of II–VI materials, where robust excitonic states justify the hope to push up the limit of temperatures, where efficient light emission from QDs is obtained. This work contains a selection of our recent experimental results in the field of the spin-related spectroscopy of individual CdTe-based quantum dots.

2. Samples and experiment

Samples studied in this work contained self-assembled quantum dots, grown mainly by the tellurium desorption method [3, 4]. A typical sample design is shown in Fig. 1.

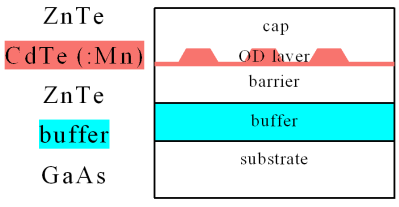

Fig. 1. Typical sample scheme.

A 4 μm CdTe buffer was grown on a GaAs substrate. Then first a ZnTe barrier was deposited, with a thickness up to 1 μm. Self-assembled quantum dots were formed from a few monolayers CdTe, deposited on the ZnTe barrier. The formation of the quantum dots was stimulated by deposition and subsequent desorption of an amorphous Te layer. Growth of an outer ZnTe barrier, typically 50 nm thick, completed the sample fabrication procedure.

Individual QD photoluminescence was used as a basic characterization tool. A special microscope [5] was immersed in superfluid He, with the sample mounted on its front surface. A typical overall spectrum is shown in Fig. 2. Because of a high density of individual QD lines in the spectrum, it was difficult to select a set of lines originating from a single QD. It was possible only in the long wavelength tail of the spectrum, where the line density was lower, as shown in Fig. 2.

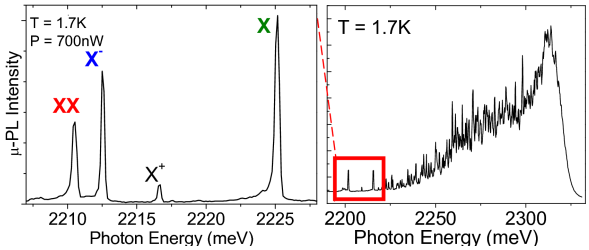

Fig. 2. Microphotoluminescence spectrum (right) of a self-assembled QD sample. A set of lines originating from a single QD visible in the blow-up on the left. They originated from neutral (X), positively (X^+) and negatively (X^-) charged excitons, and from biexcitons (XX).

Besides the standard microphotoluminescence (both cw and time-resolved), photoluminescence excitation and photoluminescence mapping have been used as experimental tools, as well as photon correlation measurements, performed in a Hanbury-Brown and Twiss detection scheme [6].

3. Charge state of the quantum dot

The charge state of a quantum dot is a crucial factor for the possibility of information recording. For example, a neutral exciton can keep its spin orientation only during its radiative lifetime, whereas a carrier left after recombination of a charged exciton can conserve a longer memory of the original trion spin orientation. The charge state dynamics of a QD was studied using photon correlation measurements. We found that the charge state variation rate depends strongly on the excitation intensity and photon energy. Under non-resonant excitation (not shown), i.e., at photon energies above the band gap of the barrier material, we found that the excitation of the quantum dots occurred predominantly via capture of single carriers — electrons or holes. As a result, e.g. X–X^- cross-correlation histograms exhibited a characteristic asymmetric shape, as opposed to symmetric autocorrelation histograms [7]. Figure 3 shows an autocorrelation histogram of charged exciton photons and two cross-correlation histograms: neutral exciton–charged exciton and biexciton–exciton ones. They were obtained under quasi-resonant excitation, using photon energies below the barrier gap. Let us note the symmetric shape of the cross-correlation histogram. It indicates that the excitation occurs predominantly via capture of entire excitons by the QD.

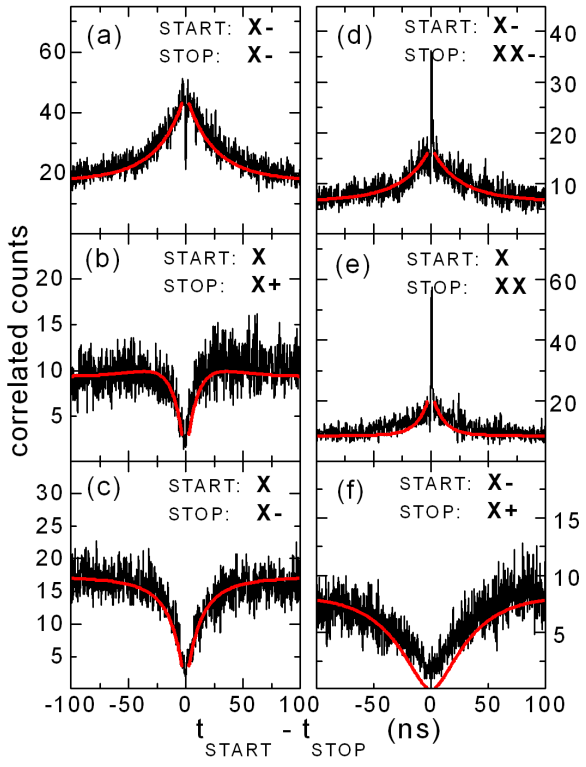

Fig. 3. Histograms of charged exciton photon autocorrelation (a) and of cross-correlations between various types of excitonic photons (b)–(f). Gray lines — rate equation model of Ref. [8]. Excitation photon energy 2.11 eV, i.e., well below the gap of ZnTe barriers. Data after Ref. [8].

The histograms contain features in two timescales. A sharp antibunching dip, below 1 ns wide (Fig. 3a–c,f), reflects single photon emission of the quantum dot: to emit a second photon, the quantum dot must be re-excited. The same sub-ns timescale applies to a bunching peak, characteristic of the biexciton–exciton cascades (Fig. 3d–e). A much wider timescale (tens of ns) corresponds to the variation of the charge state of the QD. This process is much slower, indicating that the capture of a single charge carrier (charge state variation) is much less probable than the capture of an entire exciton (re-excitation of the QD).

4. In-plane anisotropy

As shown first for the GaAs–AlGaAs system [9], an in-plane anisotropy lifts the degeneracy of the 0D confined exciton states, resulting in a doublet structure of the exciton line, known also as fine structure splitting (FSS). The components of the doublet exhibit orthogonal linear polarizations: along and perpendicularly to the anisotropy axis (Fig. 4).

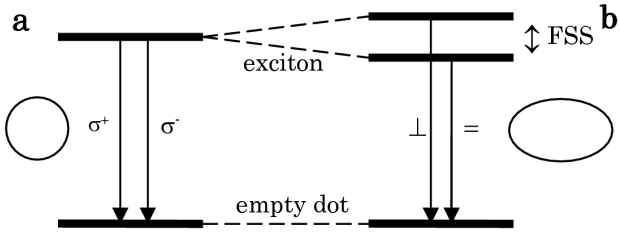

Fig. 4. Scheme of excitonic states in an isotropic (a) and anisotropic (b) quantum dot with optical transitions and their polarizations. In an isotropic QD both circular polarizations (or any of their combinations) are allowed, while in presence of anisotropy the exciton state is split by FSS and only linear polarizations are observed.

FSS is caused by electron–hole anisotropic exchange interaction. The biexciton line exhibits FSS of the same value and opposite sign, as the same exciton state represents now the final state of the optical transition. Charged exciton states are insensitive to the anisotropic exchange interaction, as they contain a pair of identical carriers in a singlet state. The anisotropy effects have been studied also in II–VI based systems, where more robust excitonic states are visible up to higher temperatures than in III–V QDs. For example in tellurides, FSS values up to hundreds μeV have been reported [8, 10, 11].

5. Information writing and read-out

One of the possibilities to encode information on a QD spin state is to use the spin of an electron left in the quantum dot after recombination of a negatively charged

exciton. The information can be then read-out by trapping a hole in the charged QD. A bright ($j = 1$) exciton thus formed will keep the memory of the original electron spin, which can be read from the polarization of the emitted photon. However, the in-plane anisotropy tends to destruct the spin memory as the pure spin states are not eigenstates of the QD exciton. This difficulty can be overcome by application of a vertical magnetic field (in the Faraday configuration). Then the Zeeman splitting restores pure spin character of the excitonic eigenstates. We showed that an electron in a single CdTe QD can keep in a vertical magnetic field a spin memory much longer than 10 ns [12].

6. Coupled QD pairs

A pair of coupled QDs is an interesting system to study spin properties. Energy and spin transfer have been demonstrated as a consequence of non-resonant horizontal coupling in a random system of self-assembled QDs [8, 13, 14]. The transfer has been revealed in photoluminescence excitation (PLE) experiments, where several emission lines, originating from the same emitting QD, were resonantly excited at the same photon energy (Fig. 5), corresponding to the neutral exciton absorption line of a neighbor QD.

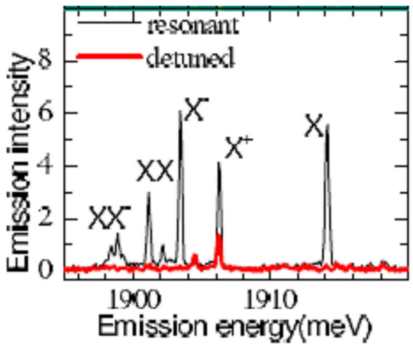

Fig. 5. Emission spectra of a single CdTe QD at two excitation energy values. The resonance causes a simultaneous enhancement of all the emission lines, corresponding to different charge states of the QD. Data after Ref. [8].

The fact that the resonance occurs at the same energy for different charge states of the emitting dot is a strong argument for the inter-dot energy transfer. The energy transfer has been shown to be accompanied by a spin transfer, visible in an optical orientation experiment. The optical orientation has been observed for all the emission lines in a vertical magnetic field, while only charged exciton lines exhibit the orientation at zero field.

7. Coherent evolution of the excitonic spin state in a QD

As already mentioned, the in-plane anisotropy lifts the spin degeneracy of the bright neutral exciton state. The exciton eigenstates can be no longer pure spin states, but their combinations corresponding to two orthogonal linear polarizations. If an exciton is created in a pure spin state, corresponding to a linear combination of linearly polarized eigenstates, the relative phase of the two components will evolve in time, resulting in an oscillatory variation of the polarization of the emitted light, starting from circular, via linear, opposite circular, perpendicular linear, back to original circular, and so on, as represented in Fig. 6.

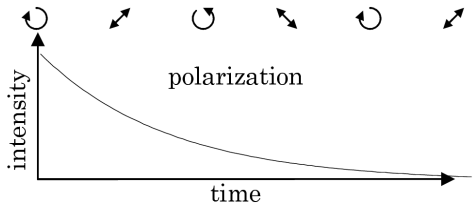

Fig. 6. Evolution of polarization and intensity of neutral exciton emission from an anisotropic QD.

This oscillation occurs with a frequency corresponding to the FSS value. At the same time, the emission intensity decays with a characteristic decay time. Combination of the oscillation and the decay results in an average polarization, different from the original one, and containing a linear component. This effect is known as circular-to-linear polarization conversion. Similarly, an excitation with a linear polarization oriented at 45° with respect to the polarizations of the eigenstates results in a linear-to-circular conversion. Both types of conversion have been experimentally detected in ensemble photoluminescence of InAs/GaAs QDs [15]. However, only small degrees of polarization have been observed in Ref. [15]. This was caused by two reasons: ensemble averaging and a large difference between the polarization oscillation period and the decay time. According to the theoretical description of the polarization conversion, the conversion efficiency may reach its maximum value of 50%, when the decay time and the oscillation period are equal. A way to overcome both difficulties has been found in Ref. [8], where polarization conversion was studied on an individual CdTe QD. A fast decay of the excitonic state was obtained by transfer of the exciton to a neighbor coupled QD. In this way, an high conversion efficiency of 40% was demonstrated, close to the theoretical maximum.

8. Single Mn atom in a QD

Interesting possibilities of using spin degrees of freedom for information recording are opened by incorporation of a single magnetic atom in a semiconductor QD. Spectroscopic studies of individual quantum dots containing single Mn ions were initiated by Besombes et al. [16] for the CdTe/ZnTe material system and by Kudelski et al. [17] for InAs/GaAs quantum dots. Figure 7 presents photoluminescence spectra of an individual CdTe QD containing a single Mn ion.

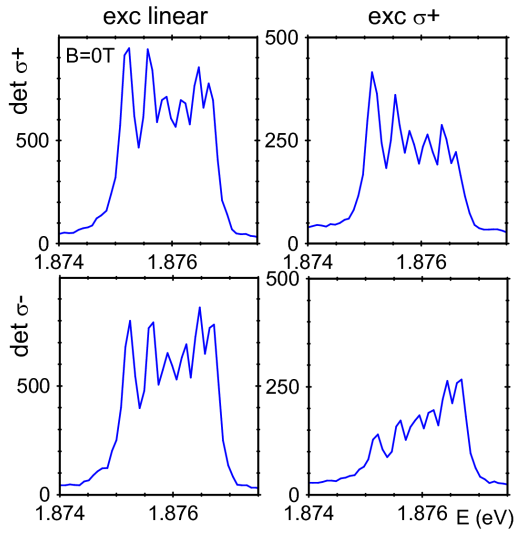

Fig. 7. Microphotoluminescence spectra of a CdTe QD with a single Mn ion, measured using indicated excitation and detection polarizations. Data after Ref. [18].

When the exciting light is linearly polarized, the spectra detected in σ^+ and σ^- polarizations are essentially identical. They exhibit a characteristic sextuplet structure, due to six possible orientations of the Mn spin with respect to the quantization direction along the growth axis. The intensities of the six components under linearly polarized excitation are equal within the experimental accuracy. Circularly polarized excitation produces spectra different in two aspects. First, the integrated intensity, detected in two circular polarizations, is different. This fact reflects optical orientation of the excitons in the QD: the probability of spin-up orientation of the recombining exciton is higher. A second new feature of the spectra lies in an inhomogeneous distribution of the intensity among the components of the sextuplet. This reflects a spin orientation of the Mn ion, occurring under influence of the circularly polarized excitation.

To achieve the optical orientation, visible in Fig. 7, we used a quasi-resonant excitation of the emitting QD via a neighbor absorbing dot, as described in Ref. [8]. This quasi-resonant excitation was also used to study spin memory of a single Mn ion [18]. It was found that the exciting light provides the main mechanism of the Mn spin relaxation. Therefore measurements of the Mn spin memory were performed in the dark. Both the intensity and the polarization of the exciting light were modulated, using acousto-optic and electro-optic modulators with temporal resolution of a few ns. The modulation cycle was following (see Fig. 8). First, σ^+ excitation was applied for a time long enough to reach quasi-equilibrium Mn spin orientation. Then the exciting light was switched off for a certain dark time, after which the excitation was restored, but in σ^- polarization. The emission intensity was measured for the lowest-energy component of the excitonic sextuplet, in σ^+ polarization (Fig. 8).

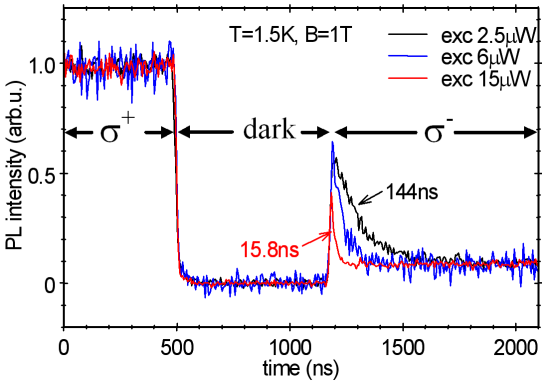

Fig. 8. Intensity of the lowest component of the excitonic emission sextuplet, measured under excitation modulated in intensity and polarization as indicated. Data after Ref. [18].

The initial value of the intensity, measured after the restoration of the photoexcitation, is smaller than the quasi-equilibrium value before the dark period. Two effects are responsible for this difference: (i) the optical orientation of the QD excitons and (ii) Mn spin orientation. Only the second one depends on the length of the dark period. Thus, we were able to study the relaxation of the Mn ion in the dark by examining the dependence of the measured initial intensity value on the length of the dark period, as shown in Fig. 9.

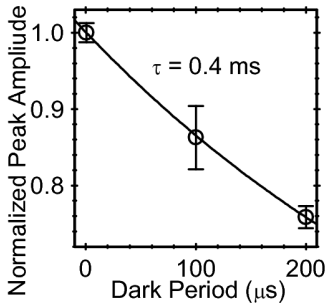

Fig. 9. Amplitude of the PL intensity peak occurring when the σ^- excitation is turned on (Fig. 8, corrected for the optical orientation of the excitons) as a function of the length of the dark period. Data after Ref. [18].

Fitting the obtained dependence with an exponential function, we were able to estimate the Mn spin relaxation time (in the dark) as 0.4 ms. The results presented in this section show that the Mn spin can be manipulated more than four orders of magnitude faster than it decays in the dark.

9. Conclusions and outlook

As shown above, telluride-based QDs represent a useful test-bed for information writing, read-out, and manipulation using QD spin states. To improve the read-out processes, more efficient extraction of the emitted photons

from the sample must be developed. New possibilities in this respect may base on placing the QDs in photonic structures. Such possibilities have been opened by recent development of distributed Bragg reflectors, lattice-matched to ZnTe barriers [19].

Acknowledgments

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References

[1] S. Cortez, O. Krebs, S. Laurent, M. Senes, X. Marie, P. Voisin, R. Ferreira, G. Bastard, J.-M. Gérard, T. Amand, *Phys. Rev. Lett.* **89**, 207401 (2002).

[2] M. Kroutvar, Y. Ducommun, D. Heiss, M. Bichler, D. Schuh, G. Abstreiter, J.J. Finley, *Nature (London)* **432**, 81 (2004).

[3] F. Tinjod, B. Gilles, S. Moehl, K. Kheng, H. Mariette, *Appl. Phys. Lett.* **82**, 4340 (2003).

[4] P. Wojnar, J. Suffczyński, K. Kowalik, A. Golnik, M. Aleszkiewicz, G. Karczewski, J. Kossut, *Nanotechnology* **19**, 235403 (2008).

[5] J. Jasny, J. Sepiol, T. Irngartinger, M. Traber, A. Renn, U.P. Wild, *Rev. Sci. Instrum.* **67**, 1425 (1996).

[6] R. Hanbury-Brown, R.Q. Twiss, *Nature* **177**, 27 (1956).

[7] J. Suffczyński, T. Kazimierczuk, M. Goryca, B. Piechal, A. Trajnerowicz, K. Kowalik, P. Kossacki, A. Golnik, K.P. Korona, M. Nawrocki, J.A. Gaj, G. Karczewski, *Phys. Rev. B* **74**, 085319 (2006).

[8] T. Kazimierczuk, J. Suffczyński, A. Golnik, J.A. Gaj, P. Kossacki, P. Wojnar, *Phys. Rev. B* **79**, 153301 (2009).

[9] D. Gammon, E.S. Snow, B.V. Shanabrook, D.S. Katzer, D. Park, *Phys. Rev. Lett.* **76**, 3005 (1996).

[10] A. Kudelski, A. Golnik, J.A. Gaj, S. Maćkowski, G. Karczewski, J. Kossut, in: *Proc. 25th Int. Conf. on Physics of Semiconductors*, Springer, Osaka 2000, p. 1249.

[11] K. Kowalik, O. Krebs, A. Golnik, J. Suffczyński, P. Wojnar, J. Kossut, J.A. Gaj, P. Voisin, *Phys. Rev. B* **75**, 195340 (2007).

[12] J. Suffczyński, K. Kowalik, T. Kazimierczuk, A. Trajnerowicz, M. Goryca, P. Kossacki, A. Golnik, M. Nawrocki, J.A. Gaj, G. Karczewski, *Phys. Rev. B* **77**, 245306 (2008).

[13] T. Kazimierczuk, J. Suffczyński, A. Golnik, J.A. Gaj, P. Wojnar, P. Kossacki, *J. Korean Phys. Soc.* **53**, 154 (2008).

[14] T. Kazimierczuk, S. Nowak, J. Suffczyński, P. Wojnar, A. Golnik, J.A. Gaj, P. Kossacki, *Acta Phys. Pol. A* **112**, 321 (2007).

[15] G.V. Astakhov, T. Kiessling, A.V. Platonov, T. Slobodskyy, S. Mahapatra, W. Ossau, G. Schmidt, K. Brunner, L.W. Molenkamp, *Phys. Rev. Lett.* **96**, 027402 (2006).

[16] L. Besombes, Y. Léger, L. Maingault, D. Ferrand, H. Mariette, J. Cibert, *Phys. Rev. Lett.* **93**, 207403 (2004).

[17] A. Kudelski, A. Lemaitre, A. Miard, P. Voisin, T.C.M. Graham, R.J. Warburton, O. Krebs, *Phys. Rev. Lett.* **99**, 247209 (2007).

[18] M. Goryca, T. Kazimierczuk, M. Nawrocki, A. Golnik, J.A. Gaj, P. Kossacki, P. Wojnar, G. Karczewski, *Phys. Rev. Lett.* **103**, 087401 (2009).

[19] W. Pacuski, C. Kruse, S. Figge, D. Hommel, *Appl. Phys. Lett.* **94**, 191108 (2009).