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Spin in CdTe/ZnTe Quantum Dot: Its Potential for Information Storage

T. Kazimierczuk

Institute of Experimental Physics, University of Warsaw, Hoża 69, 00-681 Warsaw, Poland

We present a selection of our studies on CdTe/ZnTe quantum dots considered as spin qubits. Discussed experiments are related to processes of spin reading, writing and evolution. We show that CdTe/ZnTe system is well suitable for studying effects important for optical quantum computing on single spins.

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1. Introduction

Inventions of transistor [1] and integrated circuit [2] established a close link between semiconductor physics and information processing. Rapid development of the field, aptly described by Moore's law [3], based on optimization of growth and processing techniques as well as on progress in designing complex CPU circuits.

Quantum phenomena, inevitable in nanoscale systems, have been considered a limitation to further development of classical electronics according to Moore's law. Simultaneously, quantum mechanics was recognized as an opportunity to extend the bounds of classical computational paradigm through an idea of quantum computer, described by Deutsch in 1985 [4].

Physical realization of proper building blocks for quantum computer — qubits conform to certain requirements [5] — exhibits major experimental difficulty. Many different approaches were proposed, including trapped ions [6], cavity QED systems [7], liquid state NMR [8], gated quantum dots [9], superconductors [10], crystal lattice impurities [11], and others.

Among all these possible solutions, solid-state systems are particularly promising due to relative ease of qubit manipulation, e.g., by electric, optical or magnetic fields. Prospects of future integration of qubits with existing classical electronics also constitute a significant advantage. Drawbacks of the solid state-based systems are related mainly to short coherence times limited by the interaction with the environment. The interaction with the surrounding crystal matrix may be suppressed by e.g. constraining otherwise mobile carriers inside potential well of a quantum dot.

This paper presents a selection of our studies of spins in self-assembled CdTe/ZnTe quantum dots from the point of view of quantum information processing. We will discuss three basic processes related to a single quantum dot used as a qubit, that is read-out of encoded information, writing (initialization) the qubit state and evolution of the qubit state which provides means to manipulate it.

2. Samples

2.1. Growth

Effective quantum confinement requires growing structures smaller than bulk exciton radius (6 nm in the case of CdTe). The required scale is therefore inaccessible to typical processing techniques such as electron or ion beam lithography. Instead, a bottom-up approach introduced by Stranski and Krastanov [12] is used. Under certain conditions during heteroepitaxial growth, strain originating from lattice mismatch induces a transition to 3D growth mode. Among a large variety of different material systems used in quantum dots (QD) studies such as GaAs/InAs or GaN/InGaN we selected CdTe/ZnTe heterostructures. These materials give opportunity to test basic concepts and models in relatively simple experiments, because of its well developed technology and easy visible spectral range of emission and absorption.

Studied samples were grown by molecular beam epitaxy (MBE) on GaAs substrates. A several μm thick CdTe buffer was grown to reduce negative influence of the lattice mismatched substrate material on the QDs. In the main part, samples contained ZnTe or $Cd_xZn_{1-x}Te$ barriers surrounding a few monolayers of CdTe. These CdTe layers were deposited in the atomic layer epitaxy growth mode and were precursors to the QDs. There are 2 main methods to form QDs from the CdTe layer. In the simpler one, the CdTe formation layer is annealed before growing cap layer [13]. In this case the physical situation is usually described as localization of excitons by thickness fluctuations of the CdTe quantum well.

QD with deeper confining potential can be grown using a more complex method. In this case, the process of QD formation is induced by covering the grown CdTe layer with an amorphous tellurium layer and its subsequent thermal desorption [14, 15]. As a result, a sharp transition from 2D to 3D character of the surface in the reflection high-energy electron diffraction (RHEED) image is observed, evidencing QD formation in Stranski–Krastanov-type process. The bare dot morphology was also studied by atomic force microscopy (AFM) measurements on samples whose growth had been terminated after the stage of amorphous tellurium desorption during the MBE process [15]. The QDs fabricated this way exhibit typical densities in the range 10^9–10^{10} cm^{-2} [14, 15].

2.2. Characterization

The samples were studied optically at low temperature (1.5–2 K). A specially designed microscope [16], immersed together with the sample in liquid helium, allowed us to collect photoluminescence (PL) from a small spot below 0.5 μm in diameter. This spatial selection is necessary to avoid averaging over a large QD ensemble. In most cases, we were able to distinguish a spectrally separated set of lines recognized as originating from a single QD.

A characteristic pattern of single CdTe QD emission is presented in Fig. 1a. Due to random character of carrier trapping, lines originating from different charge states are present in the same time-integrated PL spectrum.

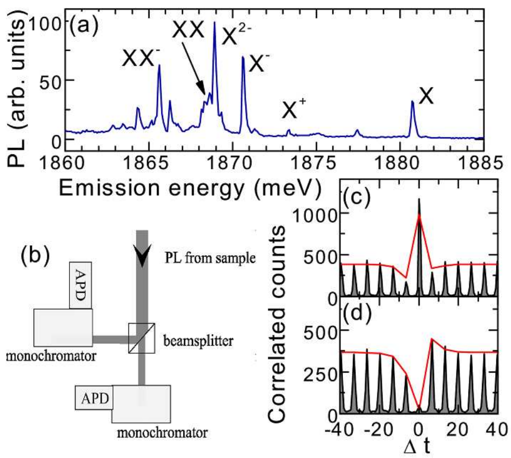

Fig. 1. (a) PL spectrum of a typical quantum dot. (b) Scheme of HBT detection setup used in correlation experiments. (c) Cascade-like correlation histogram between neutral biexciton and exciton. (d) Asymmetric correlation histogram between charged and neutral exciton.

Photon correlations between different pairs of lines were used as a versatile identification tool. The experiment was performed in a Hanbury-Brown and Twiss (HBT) setup, in which photoluminescence was measured using simultaneously two independent monochromators equipped with avalanche photodiodes (APDs) as single photon detectors (Fig. 1b). The HBT scheme, originally employed in astrophysics [17], has been widely used in studies of non-classical light emitters, e.g. single atoms [18] or molecules [19]. The simplest application of the correlation measurements is a detection of single photon sources, which are recognized by the presence of an antibunching dip in autocorrelation histograms. The antibunching dip reflects zero probability of immediate emission of two photons after one another, since some time is required to re-excite the source after emission of a photon [20].

In our studies, we measured mainly cross-correlations, i.e., correlations between photons of different energies, representing different transitions in the QD. The cross-correlation histograms reveal relations between transitions occurring in the same charge state of the QD, e.g. recombination cascades (Fig. 1c), as well as relations between transitions of different charge states as in Fig. 1d. A more detailed description of photon correlation techniques can be found in [21].

All studied QDs exhibited in-plane anisotropy. This kind of anisotropy may originate from shape asymmetry or strain resulting, e.g., from inhomogeneous QD distribution. The in-plane anisotropy influences fine structure of the QD states, e.g., it splits otherwise degenerate state of neutral exciton. As a consequence, XX and X transitions in the PL spectrum are split into doublets of linearly polarized lines. Values of anisotropic splitting of the X state vary from 50 μeV to 200 μeV, depending on the dot. The anisotropy-related linear polarization is also present for transitions XX$^-$ and X^{2-} confirming identification based on photon correlations [22].

3. Spin read-out

The optical read-out of the spin state is based on selection rules governing radiative recombination of excitonic states. The orientation of the spin of the recombining electron–hole pair is directly mapped onto polarization of emitted photon, e.g. spin up (down) are related to σ^+ (σ^-) circular polarization [23]. This correspondence can be exploited not only to measure spin state of a neutral exciton, but also indirectly to detect spin state of a single electron captured in a dot. The latter case is more interesting due to much longer lifetime, not limited by radiative processes.

A read-out of a single electron spin was realized by supplying a hole into the dot by means of non-resonant excitation and measuring the polarization of emitted photon [24]. This possibility was demonstrated in a polarization-resolved photon correlation experiment.

Two monochromators used in the experiment were tuned to observe photons from X$^-$ and X transitions, respectively. The histograms of correlated pairs count versus time distance between both photons in a pair were analyzed separately for each combination of circular polarizations of involved photons (Fig. 2b,c).

The histograms in Fig. 2b,c have their central peak strongly suppressed and exhibit an asymmetric shape, characteristic of X–X$^-$ cross-correlation. This is known to originate from the QD charge state variation under non-resonant excitation, which favors capture of single carriers instead of entire excitons [21]. The central peak of the X–X$^-$ histogram (Fig. 2b,c) represents the detection of pairs consisting of X and X$^-$ photons emitted following the same excitation pulse, therefore, its suppression reflects expected antibunching of X and X$^-$ photons similar to that observed in auto-correlation of photons emitted by any single-photon source.

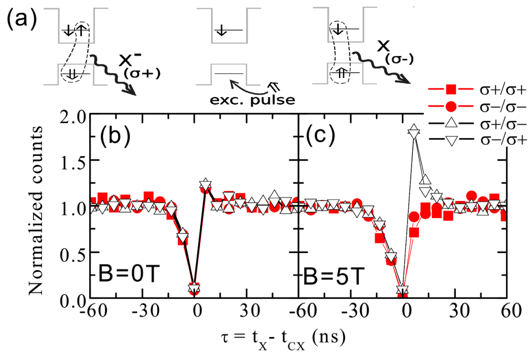

Fig. 2. (a) Scheme of consecutive recombination of charged and neutral exciton illustrating expected relation between polarizations of both photons. Cross--correlation between neutral and charged exciton emission in absence of magnetic field (b) or in a magnetic field applied in the Faraday configuration (c). Positive time corresponds to emission of neutral exciton after charged one. Different curves are related to different combinations of circular polarization of detected photons. Data from Ref. [23].

The most interesting part of measured histograms are data points on positive delay, i.e. related to detection of subsequent recombinations of charged and neutral exciton of given circular polarizations. Due to the Pauli exclusion principle, the single electron left after recombination of X^- has opposite spin to the one involved in the X^- recombination process. Consequently, unless its spin orientation is lost during exciton formation with randomly trapped hole, the second electron will recombine producing a photon with a circular polarization opposite to the first one.

Results of the experiment in magnetic field followed this prediction, as shown in Fig. 2c. However, at $B = 0$ T no X polarization was detected. This was due to QD in--plane anisotropy, which lifted the degeneracy of the X state, producing linearly polarized eigenstates. Therefore, the circular states probed in the described experiment faded out during lifetime of the neutral exciton. In case of symmetric QDs the described read-out procedure should work also in the absence of the magnetic field.

Performing the experiment with two different repetition rates, we also established that spin memory storage time in case of single trapped electron was much larger than 10 ns [23].

4. Spin writing

As it was shown in the previous section, the optical read-out process is inherently related to polarization of light — namely with circular polarization in case of canonical spin up/down base. Therefore, spin writing procedure can be considered from optical point of view as a process of optical orientation. Detection of the same (or opposite) light polarization in the emission serves here as a proof of spin polarization of an intermediate state, i.e. neutral or charged exciton.

The main difficulty in experimental realization of straight spin writing is distinguishing PL signal from much stronger scattered light of the writing laser. One of possible ways to circumvent this difficulty is to excite quasi-resonantly through other intermediate state. Experimental solutions used so far employed, e.g., excited states involving one or two LO phonons [25], or quasicontinuum of the QD excitations [26]. In case of CdTe/ZnTe QDs we tested the possibility of excitation through a neigbouring dot.

QD coupling has been extensively studied in many different systems. Both vertically [27] and horizontally [28] coupled QD pairs have been studied. The research has been focused mainly on resonant coupling of identical quantum dots, while studies of coupling between QDs with strongly different ground state energy have been less frequent [29]. In all cases the quantum dot pairs have been prepared by a suitable growth procedure aiming at positioning the corresponding dots at a controlled distance from one another. We concentrated on a single plane of self-assembled quantum dots with randomly created pairs of coupled dots [30].

The quantum dot coupling was evidenced by inter-dot excitation transfer revealed in photoluminescence excitation (PLE) experiments. In the experiment, the QDs were excited by a tunable CW dye laser. The CCD camera allowed us to record excitation wavelength dependence simultaneously for all lines in PL spectrum. The central result of the experiment were resonances presented in Fig. 3.

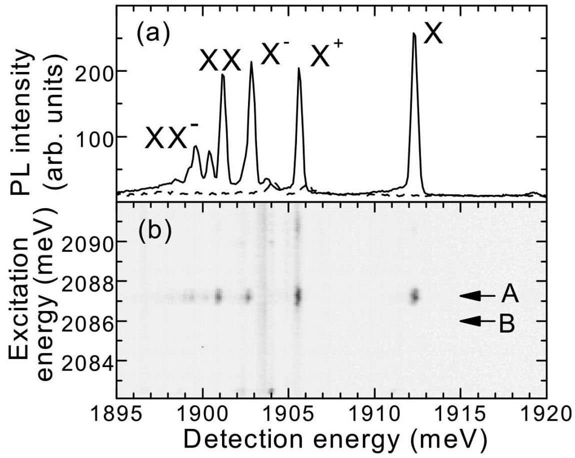

Fig. 3. (a) PL of a single QD measured under resonant excitation (solid line) and after detuning by 1 meV (dashed line). (b) Corresponding PLE map of the QD. Energies of the presented PL spectra are denoted by A and B. Data from Ref. [30].

The energies of the absorbing resonances are distributed in the range of maximum PL intensity of the QD emission band, between 100 meV and 250 meV above the corresponding emission energies. No correlation between absorption and emission energies was detected [31]. All the lines originating from each emitting QD exhibit similar resonant behavior — corresponding PLE resonance energies are equal within accuracy of tens of μeV. We interpret these facts as an evidence for the existence of two different coupled quantum dots: an absorbing- and an emitting one. In particular, the resonance occurred at the same energy for emission lines related to three different charge states, which would be very unlikely in case of one dot only.

Quasi-resonant character of the excitation affects also photon correlation histograms. Three basic types of correlation results in this regime are: cascade (e.g. XX-X), noncascade correlation between lines of the same charge state (e.g. X-X) and cross-correlation between different charge states (e.g. X-X$^-$) [30]. The main features of the histograms, i.e. bunching or antibunching at $\Delta t = 0$ are similar in case of non-resonant and quasi-resonant excitation. Difference between these two regimes is evidenced in longer timescale up to several nanoseconds. As it was shown previously, a single trapped carrier (e.g. the carrier left after recombination of a charged exciton) facilitates subsequent creation of neutral exciton, which leads to nanosecond-scale enhancement in X-X$^-$ correlation histogram. On the other hand, correlations between different charge states measured in quasi-resonant regime exhibit a symmetrical nanosecond-scale dip. This indicates clearly that main excitation mechanism supplies whole excitons instead of single carriers.

Polarization-resolved PLE measurements revealed that both coupled dots are influenced by in-plane anisotropy [30]. As in case of a single QD, the anisotropy of emitting dot was observed as splitting of emission line into two closely spaced linearly polarized components. Corresponding effect in the absorbing dot was observed as a splitting of the resonance in two components, absorbing only linearly polarized light. Interestingly, orientations of principal axes in a QD pair exhibit only weak correlation [31].

Spin writing encounters similar problems as previously discussed spin reading process — in-plane anisotropy mixes spin-up and spin-down states, producing linearly polarized eigenstates. To overcome this difficulty, we repeated PLE experiment in magnetic field applied in the Faraday configuration. The clearly visible Zeeman effect was present in coupled QDs. In field of several tesla, eigenstates of both dots turned into almost pure spin-up/down ones. A result of PLE measurements under these conditions is presented in Fig. 4a,b. Fourfold splitting of each transition is due to coexisting Zeeman splittings in both dots (Fig. 4d,e). Therefore, in this particular experiment, the orientation of circular polarization could be read from position of given maximum on the PLE map.

Successful writing of the spin state via neighbor QD depends on spin-conserving properties of inter-dot transfer.

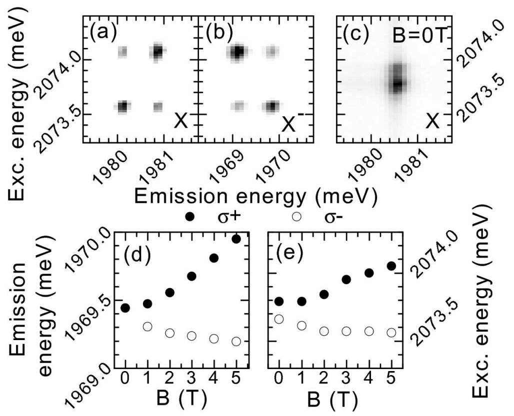

Fig. 4. (a) X and (b) X$^-$ PLE maps measured in $B = 4$ T. (c) PLE map of X without magnetic field. Zeeman effect in (d) emitting and (e) absorbing QD. Data from Ref. [30].

Experimental confirmation of this writing scheme is encoded in relative intensities of four maxima in Fig. 4a. In the magnetic field, co-polarized emission is favored over cross-polarized one. Polarization degree of X line was reduced by unpolarized contribution from XX-X cascade and yielded 20%. Polarization degree of X$^+$ line reached values close to 70% which was recognized as a more adequate measure of writing fidelity. Interesting case of spin reversal observed for X$^-$ line (Fig. 4b) is discussed in [30].

We have shown that spin writing in the canonical spin up/down base is possible in magnetic field. However, it should be noted that the profit achieved by application of the magnetic field is inherently accompanied with deteriorating of superposition state properties. Ultimate solution for the problem requires cancellation of anisotropic splitting either during the growth or by applying in-plane magnetic or electric field [32, 33].

5. Spin evolution

The main difference between classical bits and qubits is a possibility to create coherent superposition of two base states. Then different qubit states can be controlled by proper Hamiltonian engineering. We used a system of two coupled quantum dots to demonstrate evolution of spin superposition state in CdTe QD.

We used the same setup as in previously described spin writing experiment. We selected a QD pair with relatively small anisotropic splitting in the absorbing dot. Both anisotropy-split components could be excited simultaneously by a single laser. The laser was polarized at 45° with respect to absorbing QD principal axes. This configuration allowed to create (write) a linearly polarized spin state in the absorbing dot. Being a superposition of two non-degenerate eigenstates, the initial state undergoes quantum oscillations between linear states at ±45° and circular ones. The frequency of the oscillations is directly related to energy difference between two eigenstates of the absorbing dot.

Described evolution of the quantum state can be studied through emission from the other coupled dot. The quantum oscillations are accompanied by exponential transfer of occupation probability from the absorbing dot to the emitting one. In case of comparable characteristic times of decay and oscillation period, one can observe non-vanishing circular polarization of the emitted light.

Experimentally observed circular polarization followed all theoretical predictions (Fig. 5). It was observed only in the overlap range of anisotropy-split components of the resonance and required excitation at 45° with respect to absorbing dot principal axes. The sign of circular polarization reversed with changing the sign of initial linear polarization.

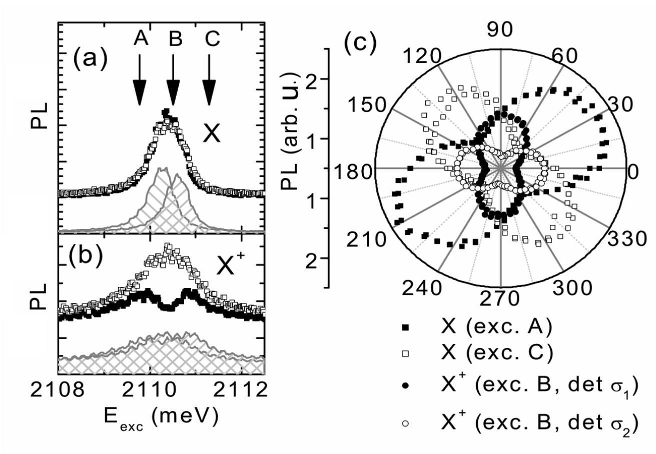

Fig. 5. Optical orientation of a QD with relatively broad resonance in circularly polarized excitation and detection. PLE spectra of (a) X and (b) X^+ transitions with excitation polarized linearly at 45° and different circular polarizations of detection (empty and full points). Gray lines correspond to excitation light polarized along principal axes of the absorbing QD (lines). The spectra were vertically shifted for clarity. (c) Angular dependence of excitation polarization required to excite both anisotropy-split components (X line excited at A and C) and to observe polarization conversion (X^+ line excited at B). Data from Ref. [30].

Very high efficiency of observed linear-to-circular polarization conversion allowed us to estimate inter-dot transfer time as several picoseconds. The inferred time is much shorter than excitonic radiative lifetime, which in turn quenches circular polarization in anisotropy-affected neutral exciton in emitting dot.

6. Summary

We study CdTe/ZnTe QDs as a medium for physical spin qubits. We highlight the prominent role of photon correlation as a convenient tool suitable for general characterization and more advanced studies such as demonstration of carrier spin read-out.

Spin read-out as well as writing is affected by in-plane anisotropy commonly present in our dots. Application of magnetic field in the Faraday configuration improves vastly writing and reading operations in the canonical spin-up/down base. On the other hand, the anisotropy can be also exploited to study coherent evolution of a single spin in the quantum dot.

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