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Recent advances in crystal optics/Avancées récentes en optique cristalline

Semiconductor heterostructures for spintronics and quantum information

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Invited Paper

Abstract

A selection of optical experiments is presented, demonstrating the utility of semiconductors in two novel areas of research: spintronics and quantum information. First we show examples of spin manipulation in semiconductor quantum wells. The light is used to generate a spin polarization and to detect it. Next we discuss application of optical methods in studies of carrier-induced ferromagnetism in quantum wells. Finally, we present examples of single quantum dot spectroscopy related to perspectives of application of quantum dots in quantum information, and, in particular, the use of photon correlation measurements as a tool to study the quantum dot excitation mechanisms. ***To cite this article: J.A. Gaj et al., C. R. Physique 8 (2007).***

Résumé

Heterostructures semiconductrices pour l'électronique de spin et l'information quantique. Un choix des expériences optiques est présenté, démontrant l'utilité des semiconducteurs dans deux domaines nouveaux de recherche : l'électronique de spin et l'information quantique. D'abord, nous montrons des exemples de manipulation de spin dans des puits quantiques. La lumière est employée pour produire la polarisation de spin et pour la détecter. Ensuite nous discutons de l'application des méthodes optiques dans les études du ferromagnétisme induit par les porteurs dans des puits quantiques. Finalement, nous présentons des exemples de spectroscopie de boites quantiques individuelles liés aux perspectives d'application des boites quantiques en information quantique, en particulier l'utilisation des mesures de corrélation de photons comme outil d'étude des mécanismes d'excitation des boites quantiques. ***Pour citer cet article : J.A. Gaj et al., C. R. Physique 8 (2007).***

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1. Introduction

Spintronics [1] and quantum information [2] are two new fields of research stimulated by application perspectives. At the same time, they open fascinating perspectives of fundamental discoveries. Spintronics aims to exploit spin of electrons instead of their charge for information transfer. Since a spin current can flow without charge displacement, hopes have been spread about a lower energy cost and a higher speed limit for the information transfer. Besides, since heat dissipation represents a serious obstacle for miniaturization of electronic components, spintronics was considered a way to push the miniaturization beyond the current limits. Although it is difficult to judge at this stage to what extent these hopes are justified, spintronics undoubtedly has opened new perspectives for both applied and basic physics. Optical methods play an important role in the development of spintronics, since light is widely used for information transfer. Spins of electrons in semiconductors can be efficiently manipulated with polarized light, therefore optical studies of semiconductors, especially of semiconductor heterostructures, represent a great potential for the development of spintronics.

Optical methods are also important for the development of quantum information, which aims at replacing the classical bit of information, possessing two logical states, by a qbit (quantum bit). Qbit states span over a whole two-dimensional space of linear combinations of basis quantum states. Quantum information methods have already proved their value in cryptography, enabling, e.g., Austrian Creditanstalt bank to execute a money transfer coded with entangled photon pairs [3]. Although semiconductors were not directly involved in the entangled photon production in that case, they undoubtedly offer tempting perspectives due to small size and power consumption of semiconductor light sources, as well as to the possibility of integration with existing electronic circuits. If the physical realization of qbits and coupling between them is sufficiently advanced, quantum computers can, hopefully, be built. Although the quantum computer represents today a distant and uncertain perspective, theoretical work shows that it may extend enormously the present computing possibilities [4].

We review here several examples of optical studies involving semiconductor heterostructures selected for their relation to spintronics and quantum information. We start from optical spin manipulation, important for these two research fields.

2. Optical spin manipulation

The physical basis of electron spin manipulation by light (OSM) has been created by the development of optical orientation, first in atomic physics [5] and later in solid state physics [6]. Optical orientation techniques provide an access to spin dynamics on picosecond time scale, using as a clock spin precession in a magnetic field. For example, in the Hanle effect [7], the spin polarization of electrons (excitons) rotates and decays under influence of a transverse magnetic field, producing a decay of circular polarization of light emitted in electron-hole or exciton recombination processes. Measurements of the Hanle effect using standard continuous wave (CW) spectroscopy give access to the spin relaxation time relative to the spin precession time. Similar advantages can be found in the resonant Faraday effect [8], where a longitudinal magnetic field induces a rotation and a decay of linear polarization of light generated by recombination of excitons in a crystal, providing also relative values of excitonic spin relaxation time. Optical orientation found an interesting application in Diluted Magnetic Semiconductors [9], where it has been used to produce long-lived photo-induced magnetization [10]. The appearance of spintronics gave a new stimulus to the OSM, and application of ultrashort laser pulse techniques opened new perspectives for this field of research, which achieved an unprecedented efficiency [11]. OSM has been demonstrated in bulk crystals, two-dimensional heterostructures [12] and quantum dots [13]. An interesting case of spin manipulation in a quantum dot is polarization conversion due to coherent evolution of excitonic doublet split by anisotropic exchange interaction [14]. We discuss below a few examples of the optical spin manipulation in semiconductor quantum wells.

2.1. Tunneling of spin-polarized excitons

Spin tunneling was studied [15] in an asymmetric double quantum well structure by means of CW excited photoluminescence (PL).

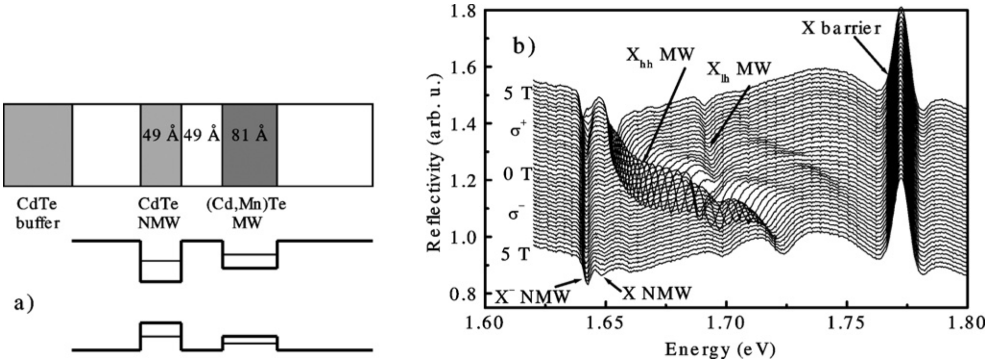

Fig. 1. (a) Structure of the sample, including a narrow non-magnetic quantum well (NMW) and a wider magnetic one (MW); (b) magnetoreflectivity spectra of the sample taken at 1.8 K in both circular polarizations. Various types of excitonic transitions in the quantum wells are indicated. After [15].

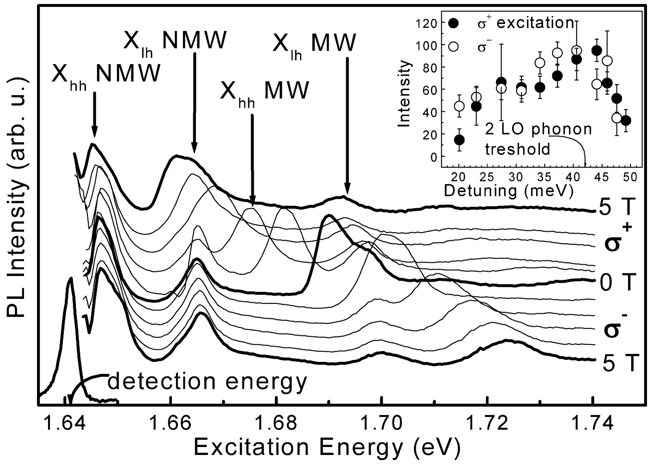

Fig. 2. Photoluminescence excitation (PLE) spectra in magnetic field up to 5 T. Insert: PLE signal versus detuning between MW heavy hole (hh) exciton and the detection energy, for two circular polarizations of exciting light. After [15].

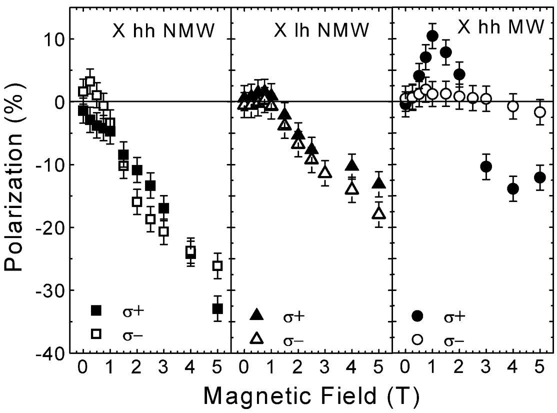

Fig. 3. Photoluminescence circular polarization, equivalent to the spin polarization of hh and lh excitons in NMW and of hh excitons in MW. After [15].

The sample (Fig. 1(a)) was grown on a (100) oriented GaAs substrate with a CdTe buffer, and contained two quantum wells embedded between (Cd,Mg)Te barriers. The wider, magnetic (Cd,Mn)Te quantum well (MW) was used as a spin aligner: The excitons, created in this well by a He-Ne laser beam, were spin-polarized by a small magnetic field through giant Zeeman effect, characteristic for the diluted magnetic (Cd,Mn)Te alloy [16]. A narrower, non-magnetic CdTe quantum well (NMW), was used to detect spin polarization, transferred from MW by a tunneling process. Reflectivity measurements in magnetic field (Fig. 1(b)) allowed us to identify several excitonic transitions in both quantum wells.

Photoluminescence excitation (PLE) spectra were measured with detection energy tuned to negatively charged exciton transition in the non-magnetic well (Fig. 2).

Magnetic field up to 5 T was used to control the detuning ΔE between the MW hh exciton and the detection energy. A maximum of the PLE signal at $\Delta E = 42$ meV (insert in Fig. 2) indicates LO phonon-assisted tunneling of entire excitons from MW to NMW. Furthermore, equal intensities for both circular polarizations of the exciting beam indicate a complete spin polarization of the excitons in MW prior to tunneling [15]. Circular polarization of the NMW charged exciton photoluminescence corresponds to spin polarization of the excitonic states after the tunneling process and capturing a carrier to form a trion (Fig. 3).

An interesting feature can be observed in Fig. 3: the recombining trions have kept a memory of the spin polarization of the excitons created by light in the MW (maximum value of 10% at 1 T) in spite of full spin relaxation in the spin aligner MW. This is caused by a heating of the MQW at σ^- polarized excitation due to the excess energy of the photocreated excitons, compared to those created by σ^+ polarized light.

2.2. Ultrafast study of spin-dependent interactions in a doped semiconductor quantum well

Pump-probe experiments represent an efficient way to manipulate spins of carriers, excitons, and magnetic ions in a semiconductor heterostructure, as shown in pioneering studies initiated in the group of D.D. Awschalom [17]. In the example shown here, interaction between excitons, carriers and magnetic ions were studied by pump-probe absorption measurements of a (Cd,Mn)Te quantum well [18,19]. The exciton–carrier interaction leads to the creation of charged exciton (trion) states, often dominating photoluminescence spectra of quantum wells [20]. This interaction is known to be spin-dependent, since the ground state of a trion includes a spin singlet pair of identical carriers—electrons or holes. The spin-dependent exciton–carrier interaction allowed us to propose an explanation of the so-called intensity stealing, observed as an attenuation of the neutral exciton absorption line accompanying an increase of the trion line [21], occurring, e.g., under influence of a carrier density increase. Fig. 4 shows an example of this phenomenon in a time-resolved pump-probe absorption measurement.

Immediately after a strong pump pulse creating a positive trion (X^+) population, a decrease of the X^+ line intensity is observed. It is accompanied by an enhancement of the exciton (X) line—we observe, in fact, a kind of inversed intensity stealing. We explain it in terms of spin-dependent screening of excitons by free carriers: when a part of the carriers becomes bound in trions created by a strong pump pulse, the carriers do not screen the excitons any more, and the exciton line intensity increases. The spin-dependent character of the exciton–carrier interaction is demonstrated in Fig. 4 by a strong dependence of the intensity variation on circular polarization of the probing light, which selects spin components of excitons and carriers. The effects are observed only in co-polarized configuration, showing that free holes of a given spin screen efficiently only excitons with opposite hole spin.

By binding in trions a part of free holes of a given spin (determined by pump beam polarization) we create a non-vanishing spin polarization of the hole gas in the quantum well. The density of unpaired holes can be measured by the difference of trion line intensities measured at opposite circular polarizations. In a time-resolved experiment we can follow the decay of thus created spin polarization [22], as shown in Fig. 5.

We determine thus the spin relaxation time of free hole gas, equal in this case 8 ps. This value is significantly smaller than the spin flip time observed under weak excitation [20]. The difference can be explained by an enhancement of carrier–carrier collisions in our case. A much slower relaxation can be observed for the Mn ion spins in the (Cd,Mn)Te quantum well. In fact, in our experiments with 100 MHz repetition frequency, we see it as a steady state effect.

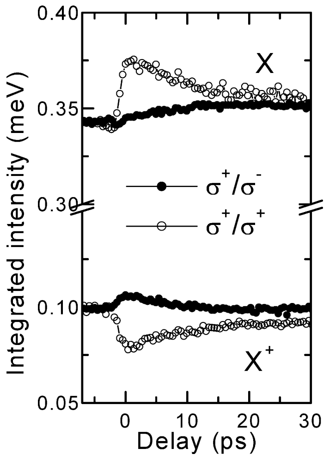

Fig. 4. Temporal evolution of exciton (X) and trion (X^+) line intensities under influence of a resonant pump pulse, tuned to X^+ energy. Pump and probe polarizations indicated for each curve. After [19].

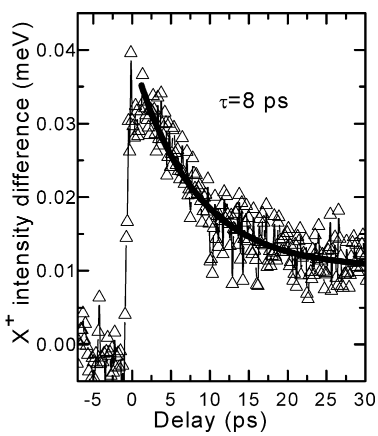

Fig. 5. Temporal evolution of difference between trion line intensities measured at opposite circular polarizations. Spectrally wide pump pulse excited resonantly both X^+ and X states. After [22].

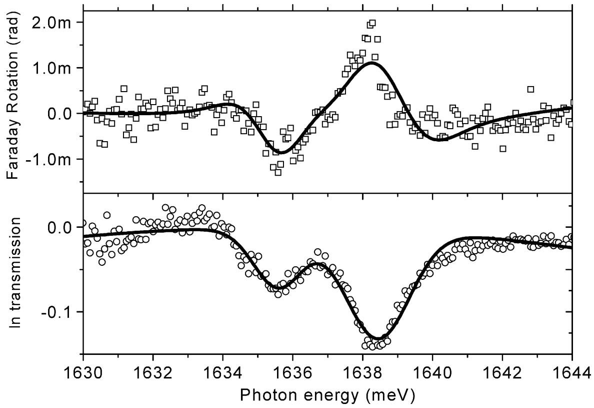

Fig. 6. Photoinduced Faraday effect in a (Cd,Mn)Te quantum well, compared with its transmission spectrum. After [18].

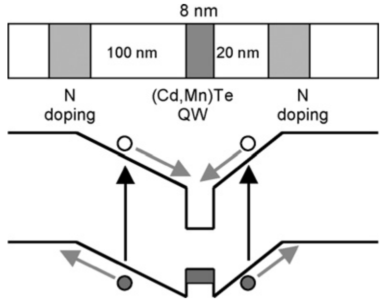

Fig. 7. Scheme of the sample with a doped (Cd,Mn)Te quantum well for optical studies of carrier-induced ferromagnetism and mechanism of carrier density control by blue illumination.

Consecutive pump pulses build up a Mn spin polarization by the spin transfer from the population of carriers and excitonic states to the Mn ions. The resulting polarization was witnessed by photoinduced Faraday effect. Fig. 6 shows a comparison of the Faraday rotation spectrum with the transmission spectrum of the sample.

A common fit of both curves with a Gaussian model allowed us to check Kramers consistency of the two spectra and to determine excitonic Zeeman splitting, which in turn gave the value of Mn spin polarization. We obtained 0.56% of the saturation value for the photo-induced Mn magnetization.

3. Spectroscopy of ferromagnetic semiconductor quantum wells

Development of the spintronics stimulates a search for room temperature ferromagnetic semiconductors, with ferromagnetic interaction between magnetic ions mediated by free carriers. Such ferromagnetism has been undoubtedly identified in several systems, including prototypical (Ga,Mn)As crystals (Curie temperatures still below 200 K); efforts to develop new materials are continued [23]. (Cd,Mn)Te p-doped quantum wells represent an interesting system for this research [24]. Although their Curie temperatures are low (a few Kelvins), (Cd,Mn)Te quantum wells represent a useful model system for studies of carrier-induced ferromagnetism. Their excellent optical quality, assuring narrow spectral lines, helps to achieve a deep insight into the physical mechanisms involved. In particular, control of the ferromagnetic transition by light and electric field has been demonstrated [25,26]. A typical scheme of a ferromagnetic (Cd,Mn)Te quantum well is shown in Fig. 7. The quantum well is modulation doped with free holes supplied by a nitrogen-containing layer placed at 20 nm from the well. A scheme of hole density control by an additional illumination is also shown in Fig. 7. Electron-hole pairs created by light in the barriers are separated by the electric field, and the electrons attracted to the quantum well compensate the confined holes.

Fig. 8(a) shows photoluminescence spectra of a quantum well at different temperatures.

At high temperature (4.2 K) a single PL line is visible. When the temperature is lowered, a splitting appears, reflecting spontaneous local magnetization. A convenient way to determine the Curie point is obtained by plotting the energy position of the PL maximum versus temperature (Fig. 8(b)). A kink is well visible on the curves obtained for carrier density 16×10^{10} cm^{-2} or more, indicating the Curie temperature values. Besides the local (domain) magnetization, optical measurements allow us to measure the macroscopic magnetization (averaged over many domains). As indicated schematically in Fig. 9(a), it can be deduced from the PL circular polarization of the integrated intensity of each of the split lines.

The PL polarization degree thus obtained is presented in Fig. 9 versus magnetic field. The plot shows magnetization saturation above 200 Oe but no measurable hysteresis. The magnetization reaches its equilibrium state within the time of the measurement (of the order of seconds).

Optical measurements allowed us also to study relaxation of the magnetization after a sudden change of external magnetic field. It has been done by applying magnetic field pulses with a small coil [27]. A small inductance of the

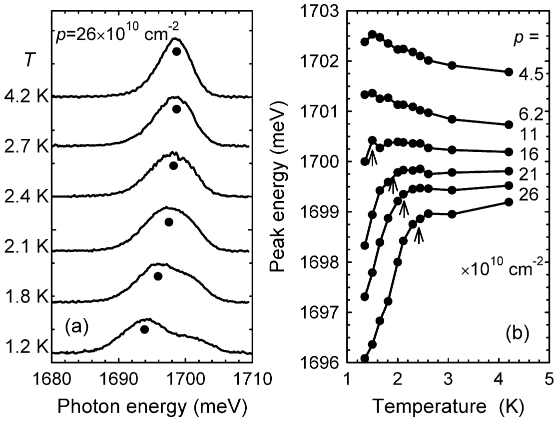

Fig. 8. (a) Photoluminescence spectra of a doped (Cd,Mn)Te quantum well, measured at indicated temperatures (b) photoluminescence peak position as a function of temperature, measured at indicated carrier sheet densities. After [26].

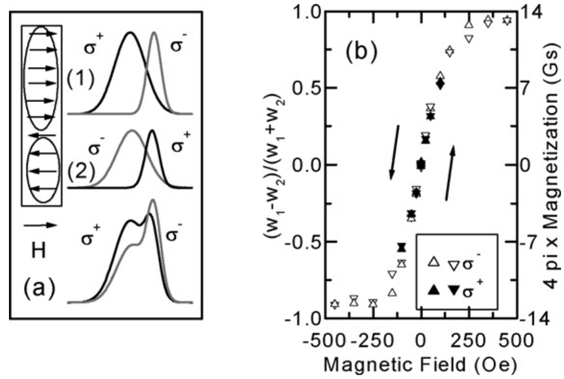

Fig. 9. (a) Schematic representation of photoluminescence averaging over domains of different orientation; (b) optically determined magnetization vs magnetic field curve; no hysteresis is visible. After [26].

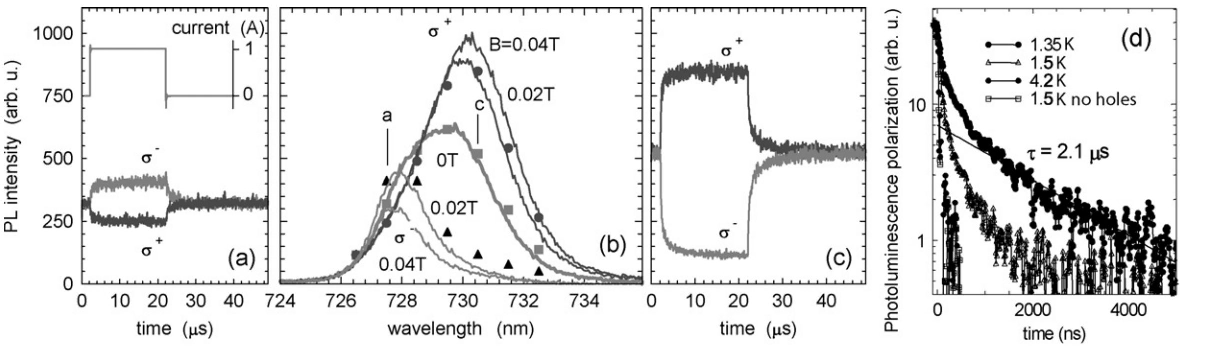

Fig. 10. (a)–(c) Time dependence of PL intensity of a doped (Cd,Mn)Te quantum well under influence of a magnetic field pulse provides information on magnetization evolution; (d) magnetization decays after magnetic field pulse show a different behavior in paramagnetic phase (4.2 K, rapid decay) than in ferromagnetic phase (1.59 K and 1.35 K, slow decays). After [27].

coil allowed us to reach magnetic field rise and decay times of about 10 ns. Setting the detection wavelength at a given point of the emission line, we registered intensity variation of opposite sign on the two slopes of the line, as a result of the Zeeman shift (Fig. 10(a)–(c)).

We focused on the magnetization relaxation at vanishing external field, measured directly after the current pulse (Fig. 10(d)). The magnetic remanence thus measured decayed with a characteristic time in the microsecond range, while quick (in tens of ns) magnetization decays were observed above the Curie temperature.

4. Semiconductor quantum dots as non-classical light sources

Semiconductor quantum dots have been attracting attention as potential sources of polarization-entangled photon pairs for quantum information applications. Photon correlation measurements have become an important experimental tool as a test of the entanglement or of single photon emission. However, correlated photon counting can be also useful to study quantum dot excitation mechanisms. We show below such an example. Correlation of photons emitted from a single quantum dot was measured in a Hanbury-Brown and Twiss setup [28]. Correlation technique was also used to measure QD PL decays [29]. In both cases a microscope immersed together with the sample in superfluid helium was used to select individual quantum dot lines. The microscope construction assured an excellent mechanical stability, enabling us to stay focused on the same quantum dot during many days of measurements.

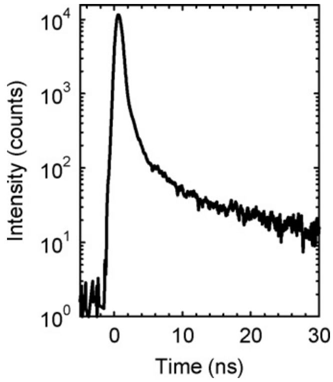

Fig. 11. Photoluminescence decay in a single CdTe/ZnTe QD under influence of a 100 ps 405 nm laser pulse. After [29].

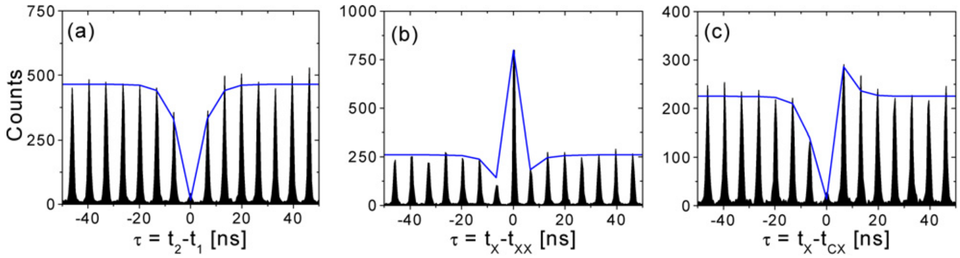

Fig. 12. Correlation histograms of photons emitted from a single CdTe/ZnTe QD under pulsed excitation: X–X autocorrelation (a), XX–X and CX–X cross-correlation (b), (c). $T = 1.8$ K, unpolarized detection. Solid line—rate equation model. After [31].

4.1. PL decay measurements

PL decays have been measured by photon counting correlated with reference signal generated by a photodiode illuminated with the exciting laser pulses. A typical PL decay curve following a pulsed excitation of a single CdTe quantum dot with ZnTe barriers by a femtosecond Ti:Sapphire laser, is shown on Fig. 11. A fast radiative decay time of about 250 ps is followed by a slow component (decay time of a few nanoseconds).

The slower component was originally interpreted as due to slow population exchange with dark exciton state [30]. However, a detailed analysis [29] indicated rather a delayed excitation process (by trapping) as a source of the slow decay component. The radiative decay times of different excitonic states (neutral exciton X, charged exciton CX, and biexciton XX) have been thus measured on a single QD.

4.2. Photon correlation measurements

After PL decay measurements, correlated photon counting was performed on the photoluminescence of the same QD [31]. Various types of second order correlation were thus measured, including X–X autocorrelation, as well as CX–X and XX–X cross-correlations (Fig. 12).

A train of pulses visible in each plot results from pulsed excitation with a temporal separation of about 6 ns. The central peak at zero delay is strongly suppressed in the X–X autocorrelation and CX–X cross-correlation histograms. This suppression reflects a small probability of emitting two photons (both X or CX–X) following the same excitation pulse. A strong enhancement of the central peak visible in the XX–X cross-correlation plot reflects an increased probability of an X photon emission following a biexciton recombination. It is worth pointing out, that the slow processes (peaks at small non-zero delays different from those in the large delay limit) indicate significant variation of the QD charge state [32]. Furthermore, significant CX–X correlated photon detection (Fig. 12(c)) represents a direct proof of the QD charge state variation. The results of the correlation measurements have been interpreted in terms of a rate equation model, describing excitation by trapping of individual carriers and excitons, and radiative recombination. We used the radiative lifetimes obtained by previous decay measurements on the dot, while three capture rates were

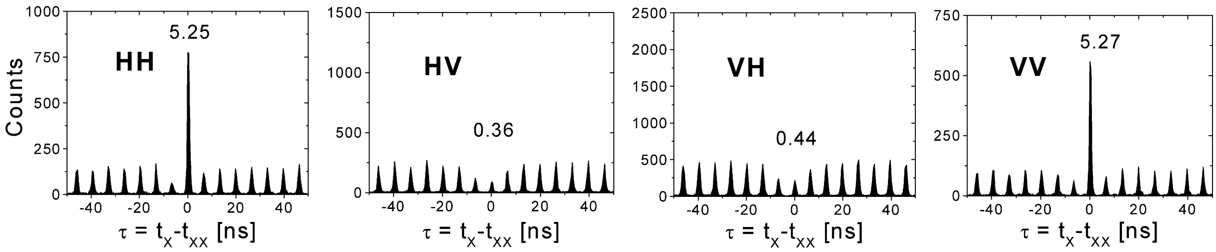

Fig. 13. XX–X cross-correlation histograms measured for a single CdTe/ZnTe QD under pulsed excitation; detection in indicated linear polarizations (H—horizontal, V—vertical) in the two arms of the HBT setup. Intensity of the central peak relative to the average of large delay peaks indicated in each plot. After [31].

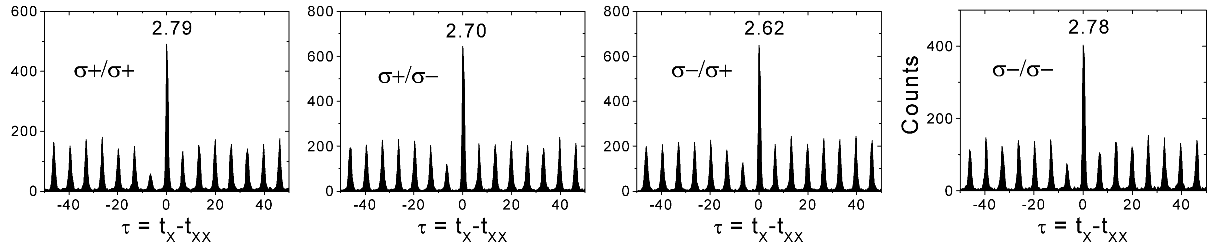

Fig. 14. XX–X cross-correlation histograms measured for a single CdTe/ZnTe QD under pulsed excitation; detection in indicated circular polarizations in the two arms of the HBT setup. Intensity of the central peak relative to the average of large delay peaks indicated in each plot. After [31].

treated as fitting parameters. An excellent description of the correlation results was obtained, as shown in Fig. 12. A relatively small value of the exciton capture rate confirms the qualitative conclusion about the importance of the single carrier capture.

We can conclude from Fig. 12 that to a good approximation single photon emission takes place after excitation pulses and that biexciton–exciton cascade is observed. A standard test of the entanglement has been also performed by polarized XX–X correlation measurements. A strong polarization correlation was observed for linear polarizations, as shown in Fig. 13.

However, no significant dependence of the XX–X correlation on circular polarization was found (Fig. 14), indicating that the photon pairs are only classically correlated and not entangled.

4.3. Role of QD anisotropy

The entanglement was not achieved probably because of anisotropy of the studied QD. This anisotropy, resulting from the dot shape, local strain or electric field, lifts the spin degeneracy of the neutral exciton through the anisotropic part of the electron-hole exchange interaction, and makes the two paths of the biexciton cascade spectrally distinguishable. Efforts were undertaken by many researchers to find a way of compensating or circumventing the QD anisotropy, to make the entanglement possible. In particular, in-plane magnetic field was used in the first successful experiment of entangled photon pair generation from InAs quantum dots with GaAs barriers [33]. In our experiments we selected in-plane electric field as a perturbation applied to compensate the QD anisotropy [34]. The electric field was generated by applying a voltage to metallic electrodes deposited on the surface of the sample. Since the Anisotropic Exchange Splitting (AES) was often smaller than the PL line width, it was measured by taking a series of PL spectra at different positions of a linear polarizer placed in the path of PL light. The AES was deduced from the oscillation amplitude of the PL line energy as a function of the polarizer angle. The difference between the line positions determined in two orthogonal polarizations on a selected InAs/GaAs QD was decreased by more than a factor of two by applying 6 V to the electrodes [34]. Fig. 15(a) shows those results plotted in a different way. Instead of the voltage we select the transition energy (average value of the two components), which represents a measure of the electric field due to the Stark shift: lower transition energy corresponds to stronger field.

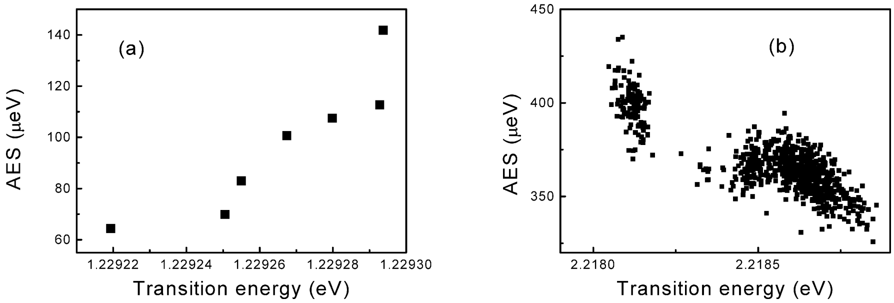

Fig. 15. (a) Anisotropic exchange splitting of a neutral exciton line of a InAs/GaAs QD, versus transition energy, Stark-shifted by applied in-plane electric field (results of [31]); (b) the same plot for a CdTe/ZnTe QD under influence of local fluctuating electric field (after [35]).

To produce entangled photon pairs we would have to compensate the AES with a great precision, corresponding to the natural linewidth. The possibility of the full compensation depends on the mechanism of the AES reduction. If the observed AES reduction comes from electron-hole separation under influence of the electric field, we cannot hope for a full compensation, since a significant electron-hole overlap is necessary to keep a reasonable emission efficiency. On the other hand, if the AES reduction is due to a wavefunction symmetry modification by the electric field, we can still hope for a full compensation of the AES in future experiments. This second version is supported by results of experiments on CdTe/ZnTe QDs under influence of fluctuating local electric fields [35]. Fig. 15(b) shows a correlation between the fluctuating transition energy measured repeatedly on the same CdTe/ZnTe QD and the AES. In contrast to the results presented on Fig. 15(a), the AES increases here with the electric field intensity, which cannot be explained by electron-hole separation in the electric field. We can therefore conclude that a symmetry modification of the exciton wavefunction takes place.

5. Conclusions

The reviewed examples of optical studies of semiconductor heterostructures show the potential of semiconductors for the development of spintronics and quantum information. In particular, spin optoelectronics can use light polarization to create and manipulate spin polarization of carriers and excitons. Optical methods can be used to characterize ferromagnetic phase in semiconductors, particularly in quantum wells containing magnetic ions and free carriers. Semiconductor quantum dots can be used to develop efficient sources of single photons on demand, as well as of correlated and entangled photon pairs. Exciton states in quantum dots can also be used to build qbits for quantum information. For all these applications a precise knowledge of quantum dot electronic states and their properties is necessary. Optical studies are perfectly suited to advance this knowledge.

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