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The Novel Multichannel Single Photon Correlations Technique Applied for the Spin Dynamics Study of a Few Mn^{2+} Ions in a CdTe/ZnTe Quantum Dot

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In this work we demonstrate a novel experimental approach to the study of single photon correlations. The introduction of the multichannel detection setup enables the simultaneous measurement of a large number of correlation functions for photons emitted from different energetic ranges. The advantages of this new approach were exploited in a detailed study of the biexciton–exciton recombination cascade in CdTe/ZnTe quantum dots doped with a few Mn^{2+} ions. The information about the dynamics of the magnetic system in the quantum dot during the lifetime of the exciton was obtained from the analysis of the correlation functions.

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1. Introduction

The optical studies of semiconductor quantum dots (QDs) provide the insight into the fundamental properties of different carrier complexes trapped in the confining potential of the dots. The large variety of methods was utilized in order to study the statical and dynamical phenomena related to the formation, evolution and recombination of multiple excitonic complexes in the QDs. Among the time-resolved studies the measurements of the photon correlations appear as a particularly convenient tool for the observation of many unique features of QDs. For example, the correlation techniques provided a way to demonstrate the application of QDs for the emission of single photons [1, 2] or the generation of the entangled photon pairs [3]. The correlation measurements performed for photons emitted from multiple excitonic complexes were also found useful for fundamental studies. They enabled the detailed description of the carriers capture and recombination dynamics [4, 5] including the identification of recombination cascades [2, 4].

In this work we present a novel approach to the single photon correlations that allowed us to extend their feasibility on the QDs doped with a few Mn^{2+} ions. The previous studies of extremely diluted magnetic QDs focused mainly on singly-doped dots [6–10]. Apart from the studies of optical properties of such structures the possibility of the optical writing and read-out of the spin state of the magnetic ion was reported [11]. However, in order to obtain a deeper insight into the properties of more complex spin systems in the QDs it is necessary to investigate the dots with a few magnetic ions. In such cases the photoluminescence (PL) lines related to the recombination of the excitonic complexes are significantly broadened due to the fluctuations of the spins of the magnetic ions. Therefore the necessity of probing various states of the magnetic system in the dot by photons of different energy requires the modification of the standard approach to the photon correlations technique.

In this paper we present the new type of experimental setup exploiting a multichannel detection module that enables the application of single photon correlations measurements to QDs containing a few Mn^{2+} ions.

2. Multichannel correlations setup

The multichannel detection module constitutes a modification of a standard Hanbury-Brown and Twiss configuration for the single photon correlations measurements (Fig. 1). Typically the PL signal is split into two arms. The detection in each arm is realized with a monochromator equipped with an avalanche photodiode at the exit. In our setup one of the photodiodes is replaced with a specially designed holder that supports a bunch of optical fibers. The spectrally resolved PL signal couples with the fibers and is transmitted to the photodiodes. The fibers are placed collinearly, therefore they enable the detection of photons from separated ranges of energy. The geometry of the fibers, their position and the monochromator's grating and focal length determine the spectral resolution of the setup. In our case the diameter of the fibers core was 90 μm with the 107 μm separation between the center of the closest cores.

In the present studies the 75 cm monochromator was used with the 1200 g/mm grating. For these parameters each diode covered the 0.3 meV energetic range with the spectral separation of 0.35 meV.

In the second arm the single diode was set to cover the 0.4 meV energetic range. The temporal resolution of the detection was limited by the response time of the avalanche diodes and was equal to about 50 ps. The presented setup enables the simultaneous measurements of

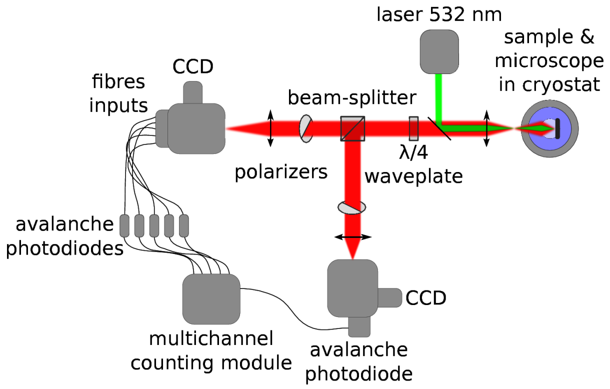

Fig. 1. The scheme of the single photons correlations setup including the multichannel detection module.

correlation functions for various choices of pairs of diodes. The maximum number of active diodes is limited by the number of channels in the single photon counting module and currently is equal to 8. In the experiments presented in this work the number of diodes actually used was 6. We applied the described setup for correlation of photons between the single diode in one arm and each of the diodes in the second arm. In such a case all the measured functions are obtained under the same experimental condition which makes a significant improvement as the photon correlation measurements usually require at least a few hours. Any instabilities in the excitation or in the detection influence the results in the same manner therefore the comparison of the information extracted from the correlation function is much more precise.

In present work the correlations measurements were performed on photons emitted from a QD with a few (2–3) Mn^{2+} ions. The sample was placed in the optical cryostat at 1.7 K and excited by the 532 nm cw diode laser. The focalization was realized with a reflective immersion microscope attached directly to the sample's surface. The diameter of the laser spot on the sample was smaller than 1 μm.

3. Samples

The sample used in the experiment was grown by molecular beam epitaxy and contained self-assembled CdTe/ZnTe QDs with Mn^{2+} ions. The concentration of Mn doping in the CdTe layer was tuned to slightly exceed the optimal value for finding singly-doped QDs [7, 8]. The isolated lines in the low-energy tail of the spectrum formed characteristic patterns of the emission lines from a single QD [4, 11–13]. Three types of dots could be observed in the presently studied sample: (1) nonmagnetic dots showing narrow excitonic lines with typical width of about 0.2 meV [12], (2) dots with a single Mn^{2+} ion exhibiting the characteristic sixfold splitting of the neutral exciton line [6] and (3) dots with broadened excitonic lines which we interpret as a fingerprint of the presence of a few Mn^{2+} ions [8]. On the basis on the information about the low concentration of Mn used in the growth we expect that dots with broad excitonic lines contain two or three Mn^{2+} ions. The PL spectrum of the QD chosen for the photon correlations measurements is presented in Fig. 2. The width of the excitonic lines is equal to about 2 meV. The positions of lines enables the preliminary identification of neutral exciton and biexciton lines which is confirmed later by the results of correlation measurements.

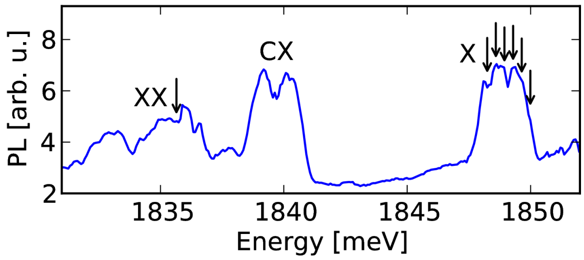

Fig. 2. The time integrated PL spectrum of the QD with a few Mn^{2+} chosen for the photon correlations measurements. The neutral exciton (X), charged exciton (CX), and the biexciton (XX) lines were preliminarily identified by the spectral position of lines. Arrows indicate spectral locations of diodes used in correlation experiment.

4. Study of the dynamics of a few Mn^{2+} ions spins in a quantum dot

The broad features observed in the time integrated PL spectrum of a QD with a few Mn^{2+} ions result from the fluctuations of the spins of the magnetic ions. The exchange interaction between the carriers trapped in the dot and the Mn^{2+} ions leads to the variation of the excitonic energy in time. Therefore the photons related to the recombination of a single excitonic complex at different energies correspond to various configurations of the spins of the Mn^{2+} ions. The time-resolved measurement of the PL signal for such dots gives the information about the temporal evolution of the magnetization of the magnetic ions.

We exploited this fact by performing the polarization-resolved correlations measurements for photons related to the recombination of the biexciton (XX) and the neutral exciton (X). In nonmagnetic dots the circular cross-polarization measurement reveals a correlation peak arising from the XX–X recombination cascade [2, 4]. The analogous measurement for circular co-polarization leads to the flat correlation signal which reflects the singlet state of the biexciton. For magnetic dots the Mn^{2+} ions might play a significant role in the cascade process. To study their influence we correlate photons emitted at the energy range around the center of the biexciton line and the photons emitted at 6 energy ranges which cover the neutral exciton line (see Fig. 2).

The correlation functions for one of the cross-polarization measurements are presented in Fig. 3. The clear peaks observed for some correlation functions are related to the XX–X cascade process. In order to quantitatively study the influence of the magnetic ions we calculate the peak area for the same temporal range from −0.5 to 1.5 ns for each correlation function.

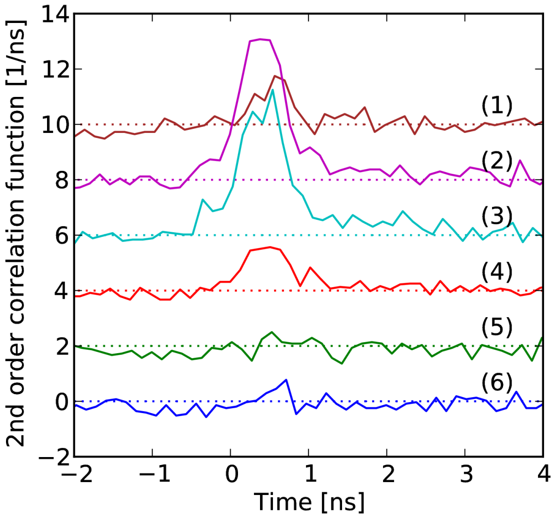

Fig. 3. The normalized correlations functions obtained for the emission from the center of the biexciton line and 6 energy ranges within the neutral exciton line in cross-polarization (σ_2/σ_1). The numbers correspond to the diodes position on X emission line according to the marked in Fig. 2 (diode no. 1 is at lowest energy, and diode no. 6 at highest). The curves are shifted vertically for clarity. The dotted lines indicate $g^{(2)}(\tau) = g^{(2)}(\infty) = 1$.

The value of the integrals are depicted in Fig. 4 for all possible configurations of circular polarizations. It is important to note here that due to the singlet state of the biexciton it does not interact with the Mn^{2+} ions system in the first-order approximation. Therefore in the XX–X cascade process the splitting caused by the interaction with the magnetic system is present only for the neutral exciton state, which is the final state of the biexciton recombination and the initial state for the exciton recombination. Therefore there is a correspondence between the Mn^{2+} spin configuration in the biexciton and the neutral exciton line in reversed order — the low energy states of the biexciton are related to the high energy state of the neutral exciton and vice versa. The integrated correlation peaks for cross-polarized detection clearly indicate that the XX–X recombination cascade occurs without the change of the spin configuration of the magnetic ions with highest probability. However we also observe significant correlation signal corresponding to the variation of the mean magnetization of the Mn^{2+} ions. This strongly indicates that the spin fluctuations occur in the timescale shorter than the neutral exciton lifetime [5].

An interesting feature might also be found in the co-polarization measurements. A significant correlation

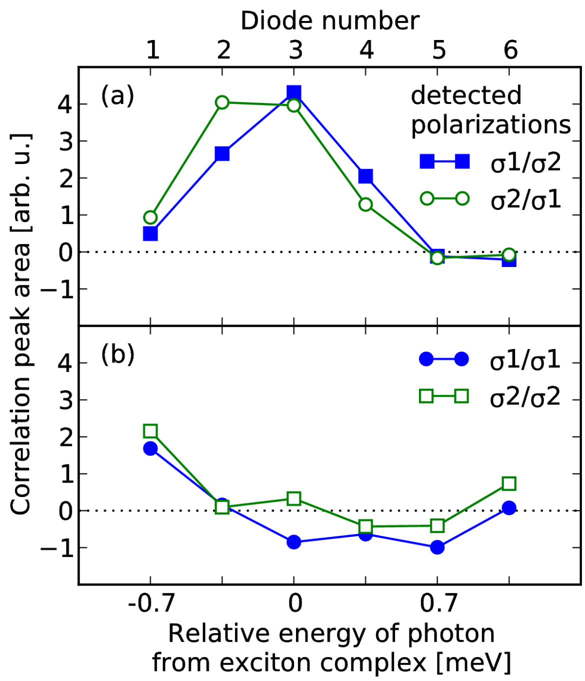

Fig. 4. The areas of the correlation peaks obtained by the integration of the correlation functions in the temporal range from −0.5 to 1.5 ns for all possible configurations of circular polarizations in detection. The upper axis shows the numbers of the diodes on exciton emission line according to the Fig. 2. The lower axis indicates the energy detuning from the center of the neutral exciton line which is assumed to be determined by the diode number 3.

peak was observed only for the lowest energy state of the neutral exciton. The observation of correlation for co-polarization requires the change of the neutral exciton's state. The co-polarization correlation was never reported for nonmagnetic dot, therefore we attribute its presence to the spin flip of the neutral exciton via the exchange interaction with the Mn^{2+} ions, which is known to be an effective relaxation channel in various magnetic heterostructures.

5. Conclusions

To summarize, we have extended the feasibility of the standard correlation setup to incorporate the multichannel detection module. Taking advantage of the possibility of simultaneous correlations of photons emitted at different energy ranges we investigated the influence of a few Mn^{2+} ions in the QD on the XX–X recombination cascade. We have found that the significant change in the mean magnetization of the Mn^{2+} ions system can be observed within the lifetime of the neutral exciton. Another finding concerned the exchange of the magnetic moment between the exciton and the magnetic ions that leads to the relaxation of the system.

Acknowledgments

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