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Resonant Excitation of CdTe/ZnTe Quantum Dot Pairs as a Tool for Spectroscopic Study of the Excitonic *p*-States

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We propose a new method of obtaining a photoluminescence spectrum of a single quantum dot in a self-assembled system of CdTe/ZnTe quantum dots. The method is based on the resonant excitation of a coupled dots pair. The comparison of the spectra in resonance and out of resonance enables the identification of a well-isolated lines related to the excitonic *p*-states. The application of the method allows the basic characterisation of a quantum dot, including the measurements of linear anisotropy, the excitation power dependence, and the analysis of the photoluminescence in the magnetic field.

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1. Introduction

The commonly applied method for the spectroscopic study of single quantum dots (QDs) in self assembled CdTe/ZnTe system is based on the combination of spatial and spectral selection. The single dots are usually selected in the low-energy tail of the broad microphotoluminescence (μPL) band. Such an approach gives an insight into several properties of the excitonic complexes. It requires only standard focalization on the spot of micrometer size, which is easy to achieve in typical μPL setup. However, the access to the emission lines related to the recombination of excitonic complexes from higher shells, like *p*-shell [1–3], are usually hindered by the broad spectrum of the other QDs ensemble. This is due to the high spectral density of QDs. Even if investigated quantum dot is selected from the low energy tail, usually only *s*-shell lines are well separated from PL lines related to other dots. Overlapping of the emission lines from different QDs might be avoided by fabrication of masks or mesas [2]. However, such methods require additional technological processing and might influence the optical properties of the selected QD, for example the polarization of the emitted light. Here we propose a new method for the observation of the emission originating from the recombination of the *p*-shell excitonic complexes, based on the resonant excitation of a QD in a system of coupled QDs [4]. This method enables the isolation of the emission lines related to *p*-states even in the presence of lines originating from other dots.

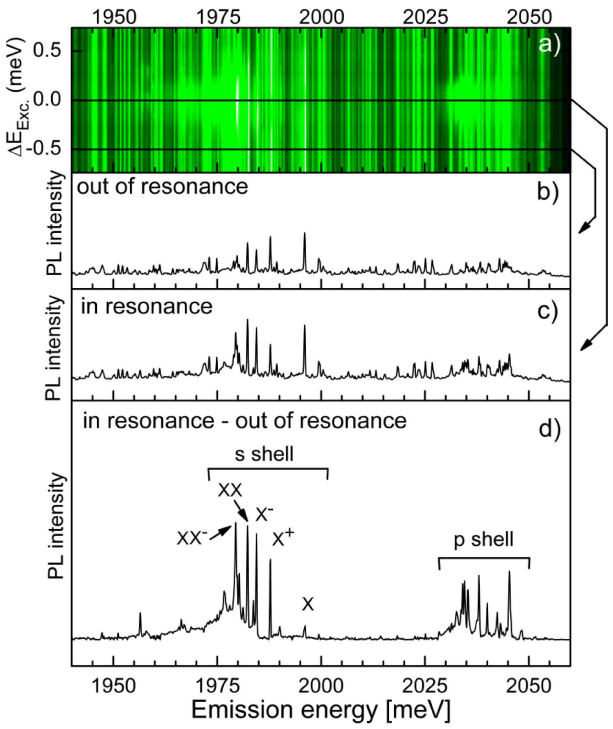

Fig. 1. The scheme presenting the new method of extracting single self-assembled CdTe/ZnTe QD spectrum from PLE (a). By subtracting spectrum out of resonance (b) from spectrum in resonance (c) the spectrum of single QD with clearly visible emission lines related to electronic *s*-shell and *p*-shell is obtained (d).

2. Method

In our experiment the self-assembled CdTe/ZnTe quantum dots were excited by a tunable dye laser in the range from 2060 meV to 2170 meV. Sharp resonances appearing in the photoluminescence excitation spectra (PLE) (Fig. 1a) indicate the presence of coupled QD pairs [4]. Subtracting spectrum out of resonance (Fig. 1b) from the spectrum in resonance (Fig. 1c) gives a spectrum of a single QD with the emission lines related to *s*-shell and *p*-shell excitonic states clearly visible (Fig. 1d).

The initial attribution of the lines to the single QD is based on the typical pattern of the emission lines for CdTe/ZnTe QDs. In particular, the pattern of *s*-shell lines for such dots is the same as the well known pattern reported in literature [4]. It involves neutral exciton (X), charged excitons (X^+, X^-) and charged biexciton (XX^-).

The attribution of the lines from p-shell group is not well established in literature, but their pattern for each of the examined QDs was similar. This, along with the results of the power dependence measurements, is a strong argument against the possibility that emission lines assigned to p-shell origin from the resonance with a third QD. Moreover the properties of the p-shell lines significantly differ from properties of s-shell lines as it is shown below.

In order to increase the sensitivity of the measurement, the out-of-resonance spectra were taken as an average from both sides of the resonance. Thus, in the differential spectrum the emission lines related to the surrounding quantum dots were quenched at least 10 times, leaving clear spectrum of the isolated dot with well separated group of lines related to the recombination from the p-shell.

3. Results

To show the usability of the new method we performed several measurements commonly used in the investigation of single QDs. We have examined a few randomly chosen dots. The identification of the emission lines is usually based on the measurements of the linear anisotropy [5] and the power dependence [6]. Our data obtained for s-shell and p-shell groups of lines are presented in Fig. 2. The linear anisotropy measurements (Fig. 2a) clearly identify s-shell lines related to exciton (X) and biexciton (XX) recombination. Charged excitons (X^-, X^+) and charged biexciton ($2X^-$) emission lines demonstrate the characteristic partial linear polarization [5]. Moreover, the well-known s-shell pattern of the self-assembled CdTe/ZnTe QDs [7, 8] additionally supports our identification of X^-, X^+ and $2X^-$ emission lines. Emission lines visible below $2X^-$ may origin from recombination of higher excitonic complexes [5]. The complete spectrum with identified lines is shown in Fig. 2b.

The linear anisotropy measurements were also performed for p-shell excitonic lines (Fig. 2c). In that case we cannot clearly identify emission lines, but significant similarities to s-shell excitonic lines were observed. Two lines (P_4, P_6) show the same polarization behaviour as the X and XX lines. The P_1 and P_2 lines act similarlz to $2X^-$ lines. Moreover P_3 and P_5 lines show a remarkable resemblance to the charged exciton emission lines.

The excitation power dependence measurements (Fig. 3) reveal a clear distinction between s-shell and p-shell electronic states. The emission intensity for lines related to s-shell (except $2X^-$) is almost linearly proportional to the excitation power. This is caused by the fact that the dominant mechanism of excitation is feeding of the QD with neutral excitons. This is characteristic for the resonant excitation regime [9]. Such exciton capture leads to different excitonic complexes in the dot after the excitation event, depending on the quasi-stable state before the capture. In particular, the presence of an electron, a hole or a dark exciton [10, 11] leads to the observation of X^-, X^+, or XX, respectively. The intensity of

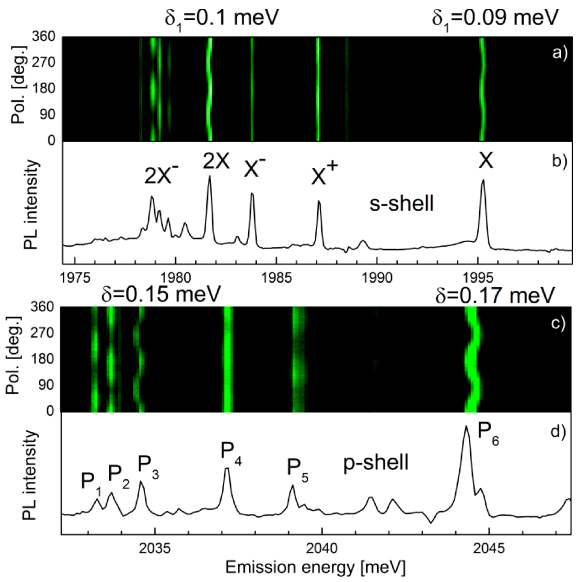

Fig. 2. The results of the linear anisotropy measurements (color-scale plot of the PL intensity vs. emission energy and the angle of the detection linear polarization) and a spectrum of identified emission lines for s-shell (a, b) and for p-shell (c, d).

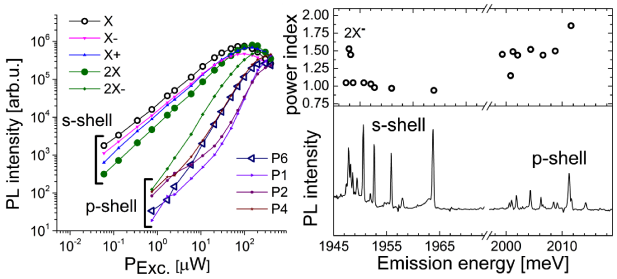

Fig. 3. Power dependence measurements results (left) and fitted power indices for electronic s-shell and p-shell emission lines (right).

emission lines related to p-shell shows the power dependence with index above one. This is related to the multi-particle capture necessary to create complexes emitting in p-shell.

One of the most important and useful application of the proposed method is the analysis of the PL spectra in the magnetic field. In such a case, the spectra are usually complex [11]. Therefore, having a pure PL related to only one dot is crucial. We demonstrate an example of the analysis of the magnetic field dependence of the PL spectrum in Fig. 4. The PLE spectra of both circular polarizations were measured for various values of magnetic field applied in the Faraday configuration and then composed to form a map. On such a map even relatively weak lines might be traced, which would not be possible for the raw PL spectra. The data in magnetic field also show significant difference between s-shell and p-shell lines. While the splitting values are comparable in

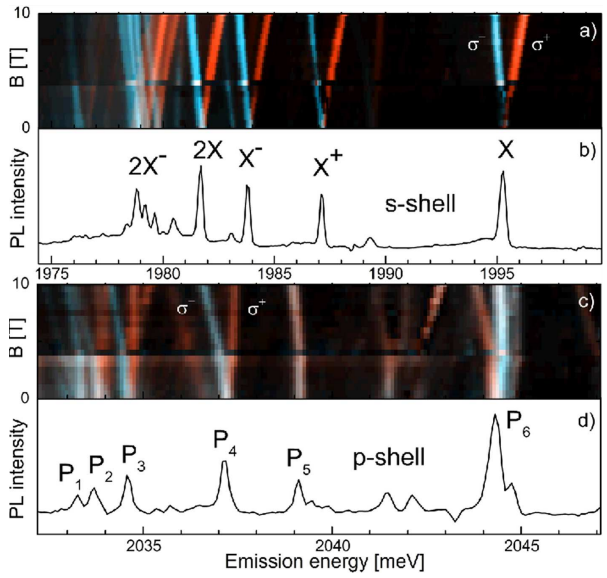

Fig. 4. The PL maps in magnetic field for s-shell (a) and p-shell lines (c), obtained by applying the subtraction procedure for every magnetic field from 0 to 10 T with step 0.5 T and for two circular polarizations of detection. Spectra without magnetic field for s-shell (b) and p-shell (d).

both groups, the diamagnetic shift seems to be larger for p-states. Although the method proposed here is limited by the necessity of finding coupled QD pair, usefulness during the measurements in the magnetic field is evident and justifies all limitations.

4. Conclusions

The new method of the observation of electronic p-shell emission lines was proposed based on the quasi-resonant excitation of a QD in a system of coupled QDs. It was shown that the common measurements of the linear anisotropy and power dependence of the excitonic lines or the scans in the magnetic field could be easily performed. The new method allows to study the recombination of p-shell excitonic complexes without any additional technological processes (e.g. fabrication of mesas). Moreover, the simple experimental setup decreases the time needed for the measurements.

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