The Fine Structure of a Triexciton in Single InAs/GaAs Quantum Dots

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Results of experimental study of multiexcitonic emission related to the p-shell of single self-assembled InAs/GaAs quantum dots are presented. Optical properties of a first emission line to appear from the p-shell of a strongly excited quantum dots are investigated using low-temperature polarization-sensitive micro-photoluminescence measurements. The emission line is attributed to the recombination of a complex of three electrons and holes confined in a dot (neutral triexciton), 3X. It is found that the emission consists of two linearly polarized components and the fine structure splitting is larger than the respective splitting of a neutral exciton. The optical anisotropy of the 3X emission is related to the anisotropy of the quantum dot localizing potential. The axis of the 3X optical anisotropy changes from dot to dot covering broad range within ±50 degrees with respect to the axis defined by the optical anisotropy of a neutral exciton (X). Possible origin of the deviation is discussed.

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1. Introduction

Semiconductor quantum dots (QDs) attract research attention due to their suitability as a basis for single photon emission for application in quantum information technology [1]. Although InAs/GaAs QDs are by far among the best understood QDs systems, there are still open questions dealing with their properties. In particular, less information is available on the QDs excited states, which are known to affect properties of QDs ground state through configuration mixing [2, 3]. It is therefore important to probe experimentally the excited states, investigating either excited states of a single exciton [4, 5] either the emission related to multiexcitonic droplets in the QD [6–8]. In this communication we report on optical properties of a triexciton (3X), which is a complex of three electrons and three holes confined in a dot. We show that the p-shell-related 3X emission line resulting from a recombination of the multiexcitonic complex to the ground state of a biexciton (2X) is strongly anisotropic. We relate the optical anisotropy to the asymmetry of the QDs confining potential.

2. Experimental procedure

The structure investigated in this work is grown on GaAs substrate by molecular beam epitaxy and contains a single layer of InAs/GaAs QDs. The indium flush technique [9] was applied and the sample was annealed after

growth (850° for 30 s) [10] in order to shift the emission energy of excitons in QDs to sensitivity range of a CCD camera. The sample was mesa-patterned in order to limit the number of dots addressed optically. The Ar$^+$ ion laser ($\lambda = 488$ nm) was used for micro-photoluminescence (μ-PL) excitation. The sample was located on a cold finger of a continuous flow cryostat at liquid helium temperature ($T = 4.2$ K). The excitation light was delivered and the PL was collected via a microscope objective (spot size around 1 μm), dispersed by using a 1 m monochromator and detected by a CCD camera. Polarization-resolved experiments were implemented using a motorized rotating half-wave plate combined with a fixed linear polariser in front of the spectrometer in order to avoid detection artifacts related to the anisotropic response function of the setup. In order to determine the energies of closely spaced emission lines, we fit their lineshape with Gaussian curves. The obtained energies of polarised emission lines follow a cosine square wave pattern. We estimated uncertainty of the energy determination as a few μeV.

3. Experimental results

Typical evolution of a single dot μ-PL spectra with increasing excitation power is presented in Fig. 1 [11].

At the lowest excitation intensity (see Fig. 1), the spectrum is dominated by two discrete lines: X and X*. Polarization-sensitive measurements revealed the fine structure splitting (FSS) of the X emission line allowed for the attribution of the X (X*) emission line to the neutral (charged) exciton in the dot. Excitation of the dot with higher power intensity results in the increase of the

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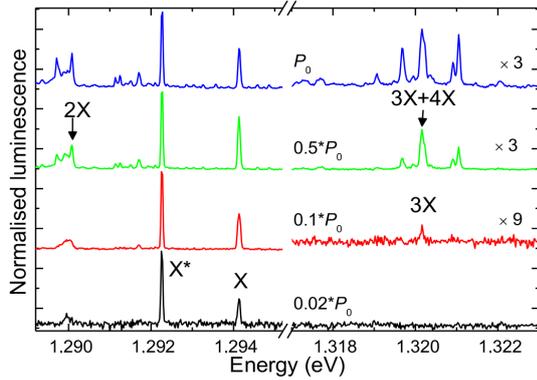* corresponding author; e-mail: maciej.molas@fuw.edu.pl

Fig. 1. Excitation-power evolution of the photoluminescence spectrum from a single InAs/GaAs dot measured at low temperature without polarization resolution.
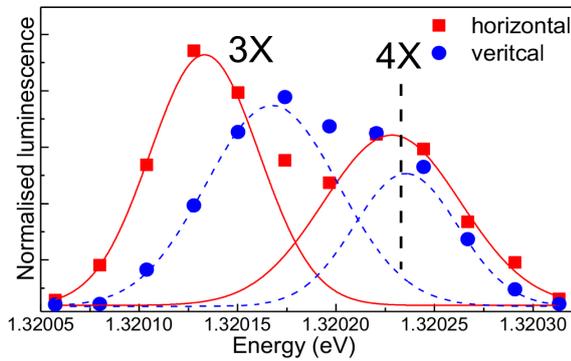

Fig. 2. The micro-PL single-dot spectrum measured in two perpendicular polarizations demonstrating the fine structure of the 3X emission.

Fig. 3. The fine structure splitting of a triexciton as a function of the fine structure splitting of an exciton in a single dot.

X and X* line intensity and in the appearance of additional emission lines. In particular the biexcitonic emission (2X) can be identified at energy of 4 meV below the X line. Simultaneously, emission lines at approximately 30 meV higher energy can be observed, which are related to the higher energy single-particle level in the dot (the p shell). The first emission line to appear in the spectrum in the p-shell-related energy range (3X) is attributed to the recombination of a triexciton to a biexciton. Following theoretical analysis presented in Ref. [12] we relate next emission lines emerging at higher excitation-power density to the recombination of a four exciton complex. Detailed analysis of the spectrum excited with the highest laser intensity is beyond the scope of the present communication. In this work we focus our attention on the 3X emission line.

More detailed analysis of the μ-PL spectrum in the 3X energy range reveals its fine structure. There are three emission lines in the energy range: two of them are polarized in perpendicular directions and one is unpolarized (see Fig. 2). In our opinion (compare Ref. [13]) the unpolarized line is due to the recombination of a four-exciton
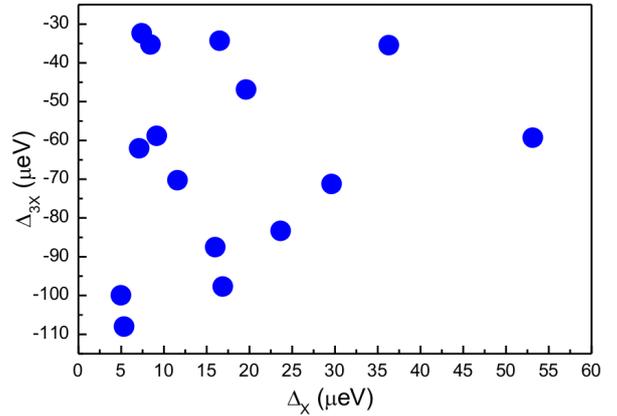
complex and will not be addressed in the following. The 3X emission line consists of two polarized components.

The energy splitting between the two polarised components of the 3X emission line is of the order of 30–110 μeV and it is significantly larger than the splitting of the neutral exciton (X) (see Fig. 3). The optical axis of the 3X emission line changes from dot to dot and is scattered in a broad range around the axis of the neutral exciton (see Fig. 3) — defined as the axis of lower energy component of the neutral exciton. The optical anisotropy axis of a neutral exciton X in turn is scattered around [110] crystallographic axis of the crystal.

4. Discussion

In a parabolic confinement approximation the single-particle p-shell is composed of two degenerate states $|p^-\rangle$ and $|p^+\rangle$ of angular momenta -1 and 1, respectively. The optically-active configuration of the triexciton is a linear combination of the states in which both electrons and holes occupy the p-shell state of the same orbital momentum [12]. If the confinement is not symmetric those states are mixed with the configurations of angular momentum ± 2 and as a result the bright 3X exciton is split [13]. The energy splitting between the bright components of the triexciton depends on the dot deformation.

The fine structure of the 3X line can be related to the fine structure of the neutral exciton emission. The larger fine structure of the 3X as compared to the splitting of the X can be understood in terms of the p- and s-shell related single-particle envelope function spatial extent. The spatial extent of the p-shell function is larger than the spatial extent of its s-shell counterpart, which may explain why it is more sensitive to the QD anisotropy. The polarisation axis of the 3X differs from that of the neutral exciton and depends on the deviation of the excitonic polarization axis from the [110] direction. In our opinion this must result from the mixing between the heavy holes and light holes in the QDs valence band [14].

For the more detailed analysis of the effect of the band--mixing on anisotropy of neutral exciton see [14]. It may be expected that the mixing is more pronounced in the annealed structures as the In/As atom intermixing during the RTA procedure releases strain in the dot and decreases the splitting between the light-hole and heavy--hole bands in the dot.

5. Conclusions

The optical anisotropy of a triexciton in single InAs/ GaAs QDs was investigated. We have found that the 3X emission line related to the recombination of a three--exciton complex to a ground state of a biexciton consists of two linearly polarized components. The energy splitting between the components is significantly larger than the splitting of the neutral exciton X. The 3X optical axis differs from the X axis, which is tentatively attributed to the effect of heavy-hole light-hole mixing in the valence band of the QDs.

Acknowledgments

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