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Spectroscopy of Indirect Excitons in Vertically Stacked CdTe Quantum Dot Structures

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We show that by means of an electric field we can tune the energy levels in vertical quantum dot pairs and study transitions related to recombination of direct and indirect excitons. With decreasing the reverse bias, we observe both the blue- and red-shifted indirect exciton transitions. Based on the band profile of our device, we conclude that the former corresponds to the recombination of the electron and hole localized in the top and the bottom dot, respectively and the latter is related to the recombination of the electron and hole localized in the bottom and the top dot, respectively.

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1. Introduction

Due to their three-dimensional confinement and possibility of easy integration into existing semiconductor devices, quantum dots (QDs) are very promising solid-state objects for building quantum optoelectronic and quantum information devices. Their functionality can be enhanced by embedding QDs in vertical field effect structures and by building structures consisting of multiple QD layers, in which new optical transitions arise. In this paper, we study samples containing two vertically stacked CdTe QD layers embedded in a *p*–*i*–Schottky structure. We present the results of photoluminescence (PL) studies of direct and indirect excitons, i.e. recombinations of an electron and a hole confined to one or two separate QDs. Obtaining a controlled coupling between two QDs is very important from the point of view of basic research and for realization of a quantum gate, the building block of a quantum computer [1, 2].

2. Experimental details

The studied samples were grown by molecular beam epitaxy on a (100)-oriented GaAs substrate. The layer sequence was the following: a 4 μm thick ZnTe buffer layer, *p*-doped with nitrogen at a level of about 10^{18} acceptors per cm^3, a 100 nm thick undoped ZnTe, two QD layers separated with a 4 or 8 nm ZnTe spacer, a 50 nm thick ZnTe cap and a 50 nm ZnMgTe blocking barrier. QDs were formed from a 2D CdTe layer, six monolayers thick by using a tellurium desorption procedure [3]. Ohmic contacts were established to the *p*-type ZnTe and a Al/Au Schottky contacts were deposited on top of the sample. Single QD pairs were accessed through 200 nm diameter shadow mask apertures produced by spin casting of polybeads.

We measured the PL signal excited below the ZnTe barrier band gap with a 532 nm laser beam focused to a 2 μm spot with a 50× microscope objective. The PL signal was collected by the same objective and detected with a monochromator with a CCD camera. Measurements were performed at 10 K as a function of a bias voltage applied to the sample.

3. Results and discussion

Typical PL spectra of a vertical single pair of CdTe QDs as a function of a bias voltage are presented in Fig. 1. Application of a bias results in an electric field given by a capacitor formula: $F = (U - U_{\mathrm{bi}})/l$, where U and U_{bi} are the applied and built-in voltage, respectively, and l is the width of the intrinsic region of our device. With increasing the electric field, PL lines shift due to the quantum confined Stark effect described by $E(F) = E(0) + pF + \beta F^2$, where $E(0)$ is the transition energy at zero electric field, p and β are dipole moment and exciton polarizability, respectively [4]. We observe two transitions: one weakly (X_1) and one strongly (X_2) shifting with bias. In the following, we present arguments which allow us to identify these transitions as direct and indirect excitons, i.e. recombinations of the electron and hole confined in the same or separate QDs, respectively.

Energies of the PL transitions as a function of the electric field are presented as points in Fig. 2. Negative F value corresponds to the electric field applied antiparallel to the growth axis of the studied sample. Fitted second order polynomials are shown as curves. A very good agreement between the fits and experimental points show that indeed the electric field is a linear function of bias. From the fits we gain access to the dipole moment p and exciton polarizability β, which for X_1 and X_2 are: $p_{\mathrm{X}_1}/e = 0.15 \pm 0.02$ Å, $\beta_{\mathrm{X}_1} = 12.1 \pm 0.03$ nm^2/V and

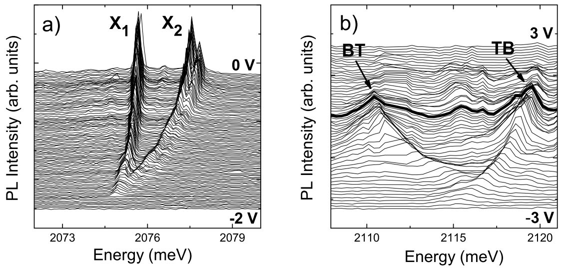

Fig. 1. PL spectra of a single pair of CdTe QDs as a function of a bias: a) from −2 V (bottom spectrum) to 0 V (top spectrum), b) from −3 V (bottom spectrum) to 3 V (top spectrum). Thick line corresponds to the spectrum at 0 V. PL transition labeled as TB (BT) corresponds to the recombination of the electron and hole localized in the top (bottom) and the bottom (top) dot, respectively. Both spectra are measured for sample with 8 nm thick ZnTe spacer layer.

$p_{X_2}/e = 1.1 \pm 0.07$ Å, $\beta_{X_2} = 27.1 \pm 0.10$ nm^2/V, respectively. Both p and β values for X_2 are greater than for X_1. Obtained p values indicate that in the case of X_2 the distance between the center of the electron and hole wave functions is larger compared to X_1. On the other hand, larger polarizability for the X_2 transition is a result of a weaker Coulomb interaction within the X_2 complex.

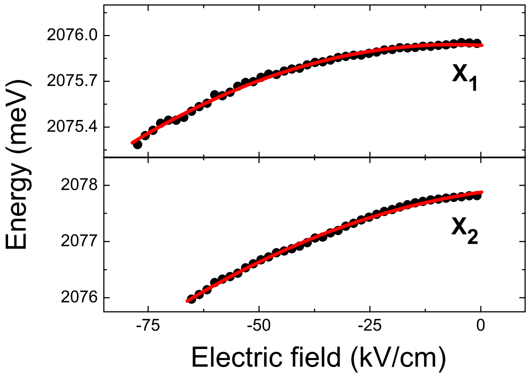

Fig. 2. Transition energies as a function of the electric field for X_1 and X_2. Lines represent fitted second order polynomials. Let us note the different vertical scales.

In order to discriminate between weakly and strongly shifting transitions we obtain p by fitting about ninety PL transitions. The dipole moment values measured for two samples with different ZnTe spacer layer thickness are shown in Fig. 3. Most of the PL transitions exhibit a dipole moment in the range of ±1 Å, indicating electron–hole separation smaller than single QD height (2 nm) and a small Stark shift. From these considerations, we conclude that these signals, including X_1, are related to the direct excitons, i.e. recombinations of the electron and hole confined to the same QD. Positive and negative p values reflect the dipole orientation [5]. $p > 0$ indicates that in absence of the electric field, the hole is located above the electron in a dot. In turn, $p < 0$ indicates that the electron is located above the hole. Let us note that both orientations are possible regardless of the exciton emission energies. Different p values may result from transitions from different QDs or different QD excitonic complexes [4, 6].

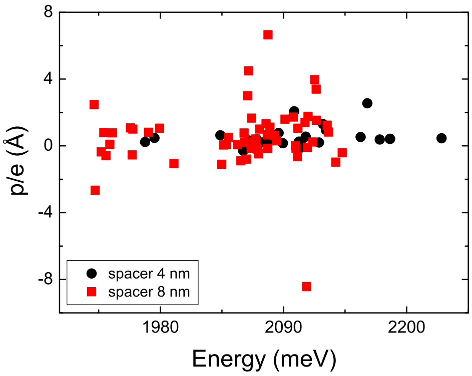

Fig. 3. Dipole moment values for direct and indirect excitons for samples containing four (circles) and eight (squares) nm thick ZnTe spacer layer.

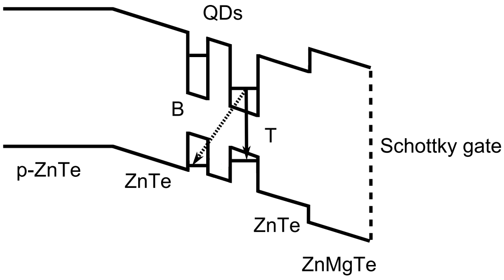

Fig. 4. Band profiles of our device for the case of zero and negative bias. The direct (solid arrow) and indirect (dashed arrow) transitions are indicated.

We identify transitions with $|p|$ substantially larger than 1 Å as indirect excitons, i.e. recombinations of the electron and hole localized in two separated QDs. We assume that for the indirect excitons the magnitude of the dipole moment, p_{ind}, should be given by the ZnTe spacer layer thickness 4 nm or 8 nm. However, p_{ind} values obtained from the experiment are about an order of magnitude smaller. This discrepancy may be caused by the external electric field being screened out by photocreated carriers in our device. Although, for dot in Fig. 1a if we assume that $p_{\mathrm{ind}} \approx 8$ nm, we get the dipole moment for the direct exciton $p_{\mathrm{dir}} = 1.1$ nm, which is a value comparable with reported p for InAs and CdTe QDs [4, 6] and also smaller than QD height. In Fig. 1b, with decreasing reverse bias we observe red- and blue-shifted transitions for which p are -8.4 ± 0.67 Å and 3.4 ± 0.49 Å, respectively. In accordance with the above conclusions we identify them as the indirect excitons. The question arises, which of these transitions is an indirect exciton with an electron in the top (T) dot and a hole in the bottom (B) dot (TB transition) and which is the BT transition. In order to provide identification, we assume that due to strain-enhanced nucleation [7], the QDs in top layer are larger than the ones in the bottom. Therefore the BT (TB) transition is expected to blue-shift (red-shift) with increasing reverse bias (confront Fig. 4). Accordingly, we identify the low and high energy transitions in Fig. 1b as BT and TB transitions, respectively. From these considerations, we conclude that the X_2 transition for dot in Fig. 1a is also TB transition, which together with X_1 are presented schematically in Fig. 4 as the dashed and solid arrows, respectively. Observation of both direct and indirect transitions suggests that electrons and holes from adjacent dots interact. Therefore, in principle, the coupling between the dots should also be possible to observe as anticrossing between the direct and indirect transitions shifting in electric field [8]. We have not observed such an anticrossing so far, most probably due to insufficient energy resolution of our setup.

4. Conclusions

We studied vertical single CdTe QD pairs embedded in the field effect structure. We observed PL transitions weakly and strongly shifting with bias, which, based on the built-in-dipole values, we identified as recombinations of the direct and indirect excitons, respectively. We find that both dipole orientations are possible, but their values are underestimated possibly due to screening effects.

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