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Emission properties of single InAs/GaAs quantum dot pairs and molecules grown in GaAs nanoholes

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Abstract. We have studied the lateral coupling between InAs/GaAs quantum dot pairs embedded in a field-effect structure. Quantum dot pairs and molecules have been identified by the correlated evolution of the Coulomb blockade features of each QD in the pair. This behaviour is largely distorted in the presence of resonant coupling states in the QD molecule. Single QD voltage evolution shows a crossover in the lineshape profile, which is associated to Spectral Diffusion processes due to residual charged environment.

1. Introduction

In the last years great efforts have been made to study self-assembled InAs Quantum Dot (QD) micro-Photoluminescence (μ PL) [1-3]. With the help of the same μ PL set up it is possible to study the quantum coupling between two neighboring dots (Quantum Dot molecules) [4]. Molecular coupling is normally identified by the anticrossing evolution of the single QD optical transitions. There have been proposed different self-assembled growth techniques to control the interdot distances and obtain an efficient coupling. They have been classified by their coupling orientation regarding growth direction. Stacked quantum dot samples are usually designed to produce vertically aligned QD molecules. In this case a field effect device is used to drive and couple the quantum levels, given that bottom and top QDs have different sizes. Molecular coupling on these vertical aligned systems has been demonstrated for electron and hole single particle states [5, 6], and even for excitons through a direct dipole-dipole interaction [7]. Laterally aligned QD molecules would be a more interesting system for quantum computing, but more difficult to produce directly by epitaxial growth techniques due to random nature of self assembling processes. The growth control of lateral QD molecules formation requires a more sophisticated technique, normally involving *in situ* etching methods [8].

In this work, we present optical signatures of single InAs/GaAs double QDs (QD pairs) grown into GaAs nanoholes under the influence of an external electric field. The *in situ* patterning procedure of the substrate by using the droplet epitaxy technique has permit to obtain low density QD ensembles with control in size and lateral configuration [9 - 11]. On these epitaxial layers, we have fabricated a lateral field effect device to study and control the coupling between dots as function of the applied

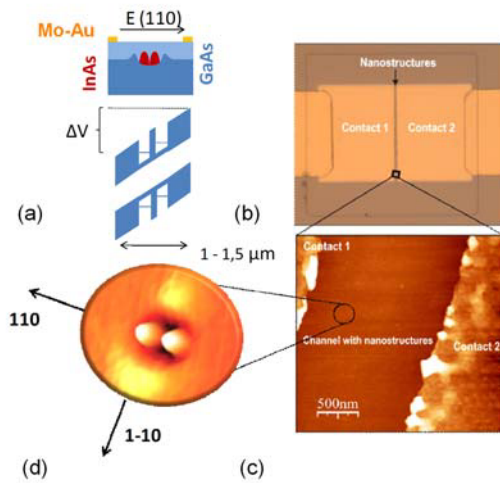


Figure 1. (a) Sample growth structure and band-edge diagram of the device. (b) Optical microscopy picture from both Mo-Au contacts. (c) AFM picture of the 1 μm gate. (d) AFM picture of one QD pair from uncapped sample.

has a mean diameter of 37 ± 4 nm and different heights with distributions centered at 5.3 ± 0.9 nm and 6.6 ± 1.6 nm, respectively. The average separation between adjacent QDs varies from practically zero at the bottom up to a value close to the QD diameter. Therefore, some QD pairs could exhibit a non negligible molecular coupling, especially for electrons. With the aim to tune the electronic levels at each QD and determine the quantum molecular coupling, if existing, an Mo-Au layer have been deposited on top of the sample surface and a 1 μm gap was opened defining the two lateral gates to apply an electric field parallel to the QD pair axis ([110] direction), as illustrated in Fig. 1.

The emission properties at the single QD level have been investigated by using a fiber based confocal microscope with a diffraction limited spot size ($\sim 1\mu\text{m}$) running at liquid Helium temperature. A Ti:Sapphire laser operated at 775 nm is used as excitation source in pulsed regime (76 MHz of repetition rate). The collected light have been dispersed by 0.3 m focal length double monochromator and detected by Si back-illuminated cooled CCD. Additionally, a programmable voltage source has been used to apply the desired voltage to the Au-Cr gate.

3. Results and discussion:

3.1. Single QD voltage sweep:

Although the QD pair occupancy at the nanoholes is about 98% [11], we have identified several examples corresponding to only one single QD nucleating at the patterned nanohole. The triplet state from a double negative charged exciton (X_T^{-2}) is recognised as the more intense peak at zero bias. This assignation is established following our previous results on these systems [9]. We have explained this negatively charged configuration by the electronic transfer from arsenic vacancies in the QD surroundings due to an incomplete Ga droplet crystallization. When performing a negative voltage sweep an abrupt change is observed in the lineshape of the X_T^{-2} optical transition, as shown in Figure 2.a. The two observed limiting situations are shown in Fig. 2.b: two narrow Lorentzian lines (limited by the spectral resolution) are observed for voltages more negative than -3.4 V, whereas a broader line that exhibits a more complicated lineshape is observed for voltages less negative than -3.2 V, similar to the case described in Ref. 12. Furthermore, an energy shift of the PL peak, $\Delta E_0 \sim 0.5$ meV (see the inset in Fig 2.a), appears between both situations. Let remember that tuning the QD charge by biasing field effect structures produce abrupt steps in emission energy [1]. Therefore, we suggest that both

bias. We have found three optical differenced regimes presumably associated to morphological differences between the QDs of the pair and the coupling strength between them.

2. Sample and Experimental Set-Up:

The sample was grown by Droplet epitaxy on a 0.5 μm thick GaAs buffer layer. During the first growth process it was generated a density of 2.10^8 nanoholes/ cm^2 with an elongated structure along [110] direction. After this nanohole formation process 1.5 monolayers of InAs was deposited at an As_2 pressure of 10^{-6} Torr for the QDs growth. Above the QDs a 100 nm thick capping layer was grown. More detailed information about sample growth mechanism and parameters can be found at [11]. Figure 1 shows the AFM image of a QD pair grown by the described procedure. Each QD in the pair

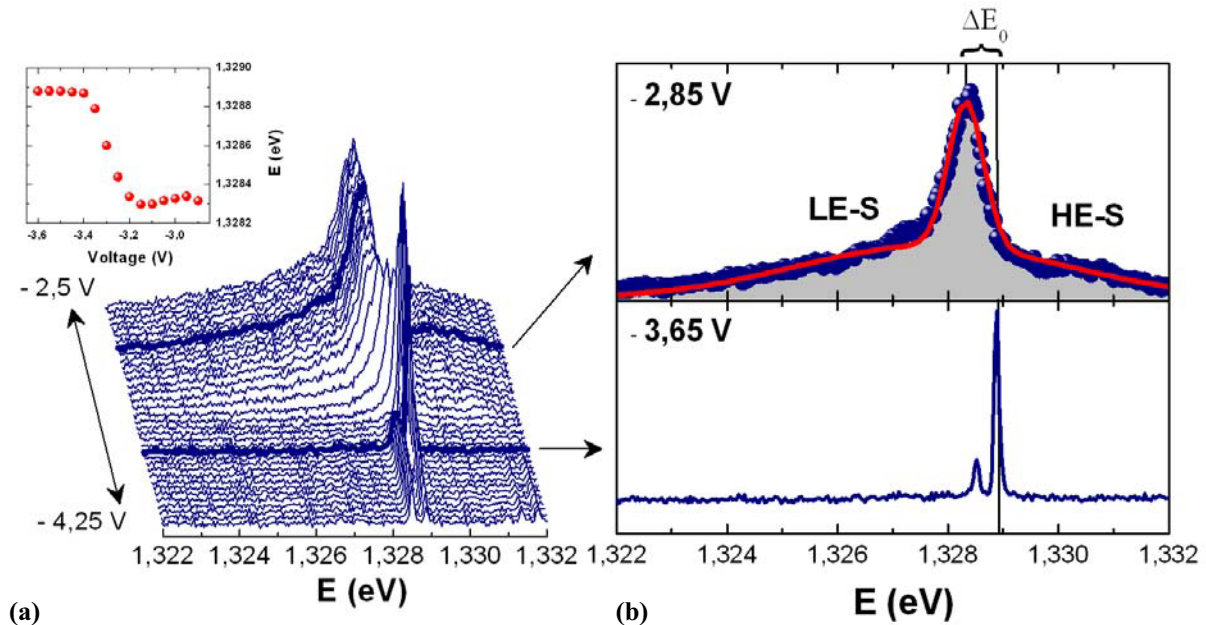


Figure 2. (a) Voltage sweep from -2.5 to 4.25 V for a single QD. Inset: Peak energy crossover of the resonance. (b) Transition lineshape at two different voltages.

changes in lineshape and energy transition are due to the Spectral Diffusion (SD) effect, associated to the fluctuating electric field of the QD charge surroundings. A similar broadening due to SD effect has been explained by the slow modulation limit of the motional narrowing effect [13], which in our case we assume to arise from local electric field fluctuations [14]. When a sufficiently negative voltage is applied between the gates, the electric field is high enough to empty the QD surroundings from residual charges, and optical transition recovers its original linewidth. However, the existence of low and high energy sidebands (LE-S and HE-S in figure 2. b) is normally ascribed to the emission and absorption of acoustic phonons [12]. We need more experimental evaluation to confirm the origin of this sideband in our system.

3.2. QD pair voltage sweeps:

3.2.1. Decoupling signatures. Figure 3.a shows a gray-scale plot of the photoluminescence versus gate voltage for a single QD pair. We identify neutral (X^0), singly charged (X^{-1}) and doubly charged (X^{-2}) excitonic features from two different QDs (QDA and QDB). This identification comes from identical procedure at similar works, which evaluate the Coulomb blockade and Pauli blocking on QD states [1]. It is not detected any sign of molecular coupling between both QD states, as their optical transitions follows the expected Quantum Confined Stark effect for isolated QDs [1].

3.2.2. Coupling signatures. Figure 3.b shows similar plot for another QD pair. In this case, the excitonic features are not easily identified as there are several peak assignment possibilities. However, following our previous discussion it is possible to identify neutral (X^0 group), single charged (X^{-1} group), and doubly charged (X^{-2} group) optical resonances sets. However, we do not have a clear explanation about the origin and causes of every line in each set. The more interesting feature of this plot comes from the asymmetric evolution of the Stark shift at positive ($\sim +2$ V) and negative (~ -5 V) voltages. It is reasonable to expect an asymmetrical evolution in molecular coupled systems, as far as the QD pair (at the same nanohole in our case) have slightly different sizes. In this case, it is possible to bring electron or hole levels into resonance by applying a negative or positive voltage, depending on the relative alignment of the electronic levels with the gate voltage. Moreover, conduction and valence bands have different band offsets, giving rise to different absolute resonant voltages for each

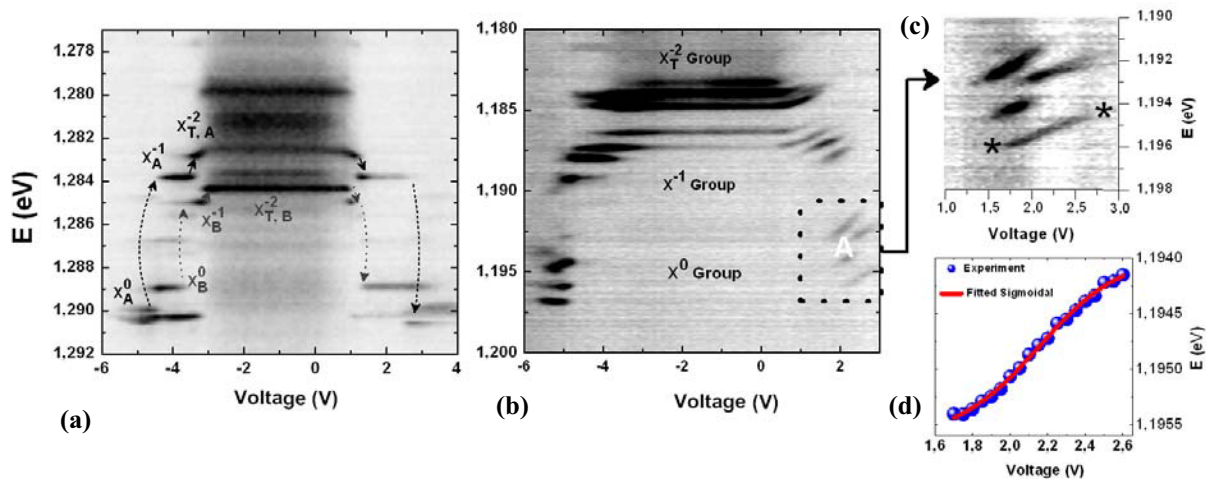


Figure 3. (a) Single QD pair voltage sweep from decoupled system. (b) Single QD pair voltage sweep from coupled system. (c) Zoom of figure 3.b. (d) sigmoidal fitting to the transition between asterisks.

carrier level. Clear anticrossing features are hardly detected in lateral quantum dot molecules [4]. However, it has been identified molecular coupling on these systems by an anomalous excitonic Stark shift evolution [8]. A zoom of the black dotted area in Fig. 3.b is plotted in 3.c, where we can observe a sigmoidal behaviour with the applied voltage between 1.7 and 2.6 V (range marked by asterisks in Fig.3.c). As better observed in Fig. 3.d, the PL peak energy of this optical transition follows a sigmoidal Stark shift variation. We suggest that both the observed asymmetrical excitonic features and the sigmoidal Stark shift evolution could be optical signatures of molecular QD coupling in our system (in some QD pairs where sizes and/or dot distance enables it).

Conclusions

In summary, we have studied the μ PL emission properties of single QD and QD pairs grown by Droplet epitaxy as a function of an external electric field. We have analyzed the lineshape of single QD optical transitions at the crossover between narrow (Lorentzian) to a broader and more complicated profile, which has been associated to a Spectral Diffusion mechanism. Molecular coupling on QD pairs has been evaluated by μ PL voltage sweeps. We have found optical signatures from decoupled QD pairs, but also to some QD pair exhibiting molecular coupling. It has been deduced from its asymmetrical behaviour with respect to the applied voltage and the sigmoidal Stark shift evolution.

Acknowledgments

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