Spectroscopy of a single semiconductor quantum dot at negative and positive discrete charge states

D.V. Regelman[1*], E. Dekel[1], D. Gershoni[1], E.Ehrenfreund[1], W.V. Schoenfeld[2], P.M. Petroff[2]

[1] Solid State Institute and Physics Department, Technion- Israel Institute of Technology, Haifa, 32000, Israel
[2] Materials Department, University of California, Santa Barbara, CA 93106 USA

Abstract We study optically single self-assembled quantum dots embedded within the wide quantum well of a mixed type quantum structure. We compare the steady state and pulsed photoluminescence spectra of these dots to those of previously studied "regular" dots. We unambiguously identify experimentally emission from various discrete charge state of the dots. We provide means for optically tune the charge state of the dot, both negatively and **positively**. Our observations are used to accurately determine the asymmetry between the quantum dots' confined electron and hole envelope wavefunctions.

1 Introduction

Optical studies of semiconductor quantum dots (QDs) have been a subject of very intensive recent investigations. It has been experimentally and theoretically established that the number of carriers which occupy a photoexcited QD greatly affect its photoluminescence (PL) spectrum.[1] In spite of its neutral nature, optical spectroscopy has very recently proved to be a useful means for investigating and preparing charged QD systems.[2,3] We report here on continuous wave (cw) and pulsed optical PL spectroscopy of single self-assembled QDs (SAQDs) embedded within a mixed type quantum well (QW) structure.[4] This specific design, which facilitates charge separation by optical means,[5] is used here to tune the charge state of the QD under study.

2 Experiment

Two samples were studied. Sample A consists of a layer of low density In(Ga)As SAQDs, embedded within the wider of a two mixed type coupled GaAs QWs, separated by a thin AlAs barrier layer.[4]. Sample B, which is used here as a control sample, consists of a similarly prepared SAQDs layer embedded within a thick layer of GaAs.[6]

We spatially, spectrally and temporally resolve the PL emission from single SAQDs in both samples using a variable temperature confocal microscope setup. The setup is described in detail elsewhere.[7]

In Fig. 1a (1b) we present the PL spectra from sample A(B) for various cw excitation powers at photon energy of 1.75 eV. In Fig. 1c (1d) we present the temporally integrated PL spectra from sample A(B) for various picosecond pulsed excitation powers by the same photon energy. The repetition rate of our Ti:sapphire laser which was used for pulsed excitation is 78MHz.

In Fig. 1d (1e) we present for comparison model simulations of the pulsed excitation PL spectra for sample A(B) as explained below.

3 Discussion

In previous works[6-8] we have established that the evolution of the cw PL emission from an optically excited SAQD is due to the recombination of increasingly higher orders of neutral multiexcitons. Characteristically, with the increase in power, satellite spectral lines appear to the lower energy side of the first observed line (X_0 in Fig. 1b) and higher energy spectral group of lines (B) emerges above the first observed group (A). This spectral red shift is explained in terms of the exchange energies between the increasing numbers of electron-hole pairs within the neutral SAQD. At yet higher excitation powers, all the observed discrete PL lines at their appearance order, undergo a cycle in which their PL intensity first increases, then reaches maximum and saturates, and eventually significantly weakens. Consequently, the various groups of spectral lines (A and B in Fig. 1b) seem to be "red shifted" with the increase in excitation power. This behavior is best described by a set of coupled rate equations[6], which give the probabilities of finding the photoexcited QD occupied by a given number of e-h pairs (multiexciton order N_x). At high excitation power the probability to find the QD with small N_x vanishes and thereby the PL lines observed at low powers disappear. This is no longer true when pulse excitation is used. In this situation with the increase in excitation power, after reaching saturation, the intensity of the PL of the various spectral lines remains constant and does not decrease with further increase in the excitation power (Fig. 1c). In this case the radiative recombination process is sequential. Therefore, all the multiexcitons which are smaller than the initially photogenerated number of e-h pairs ($< N_x >$) contribute to the temporally integrated PL spectrum. This is true, as long as the pulse repetition rate is slow such that all the photogenerated pairs recombine before the next pulse arrives.

In sample A, the SAQDs are initially charged with electrons. These electrons accumulate in the SAQDs due to the special design of the sample which facilitates efficient hoping transport from residual N-type impurities. With the increase in the excitation power the SAQDs are first photodepleted, since the positively charged traps

* *email:* dany@ssrc.technion.ac.il

efficiently capture photogenerated electrons while holes diffuse preferentially to the SAQDs.[3] The recombination of an e-h pair in the presence of a decreasing number of electrons within the SAQD reveals itself in a series of small lines to the lower energy side of the PL line due to recombination in a neutral SAQD (line X_0 in Fig 1a). The higher the excitation power is, the less charged the dot is and consequently the PL line is closer to the X_0 line. The intensity ratio between the saturated PL lines from the dot in the presence of charge and that from the neutral SAQD is roughly an order of magnitude. This ratio must be equal to the ratio between the rate by which the photexcited electrons leave the impurities and the SAQD intrinsic radiative rate. In a recent work we showed that the later amounts to 5 ns.[6]. It follows that the lifetime associated with the trapped photoexcited electrons is few times longer than the time between sequential pulses. Therefore, under pulsed excitation, PL lines from charged states saturate with increasing the excitation power and lose their intensity as the power is further increased. At the same time the PL from neutral states, saturates and remains constant (see Fig. 1d). This behavior provides an unambiguous experimental tool for the distinction between the two PL processes.

Comparison between the PL spectra of sample A and sample B reveals yet another distinctive difference. With increasing the excitation power satellite PL lines appear to the i higher energy side of the neutral X_0 line of sample A. These lines can now be unambiguously identified as resulting from recombination from QDs that are **positively** charged.

The fact that PL from negatively (positively) charged QD is lower (higher) in energy than the PL from neutral QD is reported here, to the best of our knowledge, for the first time. This phenomenon can be straightforwardly explained by solving the confined few carriers problem,[9] with different confining potentials, for electrons and for holes. The experimental results that we obtained are best fitted by our model,[6-8] as demonstrated in Fig. 1d, when a 15% smaller hole envelope wavefunction is assumed. Intuitively it means that the energy associated with the increase Coulombic repulsion due to the additional positive holes is greater than the additional exchange energy and the energy associated with the electrons-hole attraction.

4 Summary

We report on the first observation of photoluminescence from a positively charged single quantum dot. Our experimental observations strongly suggest that in self assembled quantum dots holes are better confined than electrons.

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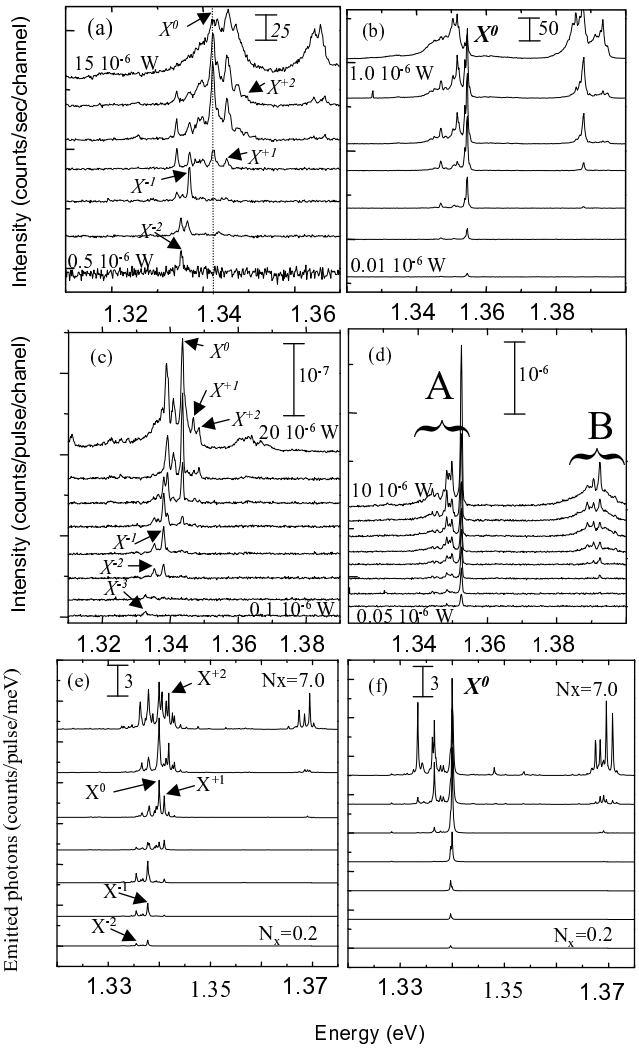

Fig. 1 a(b) [c(d)] PL spectra from sample A(B) for various cw [pulsed] excitation powers. d(e) model simulations of the pulsed excitation PL spectra from sample A(B) for various initial avarage numbers of e-h pairs $< N_x >$.

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