MULTI-EXCITON EMISSION IN QUANTUM DOTS: SPECTROSCOPIC EVIDENCE FOR A FAST EXCITON THERMALIZATION

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We apply low temperature diffraction limited confocal optical microscopy to spatially resolve, and spectroscopically study the multi-exciton emission from a **single** self assembled semiconductor quantum dot. The sharp spectral lines, which are due to optical transitions between discrete multiexcitonic states confined in the dot, depend sublinearly on the excitation power. We quantitatively explain this behavior assuming full exciton thermalization.

1 Introduction

Quantum dots (QD), in which carriers are confined to a size smaller than their de-Broglie wavelength, give a unique opportunity to study in detail the problem of "few body" interactions in restricted geometry [1]. In a recent study [2], we utilized the diffraction limited confocal optical spectroscopy technique to study a single self-assembled QD (SAQD). We showed that multiple sharp lines, as well as broad spectral bands, are the result of optical transitions between a few electron-hole pair (multi-exciton) neutral states within a single dot. Using a simple few body model we explained the measured spectral shape and transition energies.

Here, we study the emission from a single SAQD, both by CW and pulsed excitations. We present the different excitation power dependence of the various emission lines, and we show that the data can be fully explained assuming that thermalization occurs much faster than radiative recombination.

2 Results and discussion

The InAs SAQDs used here are described in detail previously [2]. Using a x100 in-situ microscope objective for 0.5 μm spatially resolved confocal spectroscopy, we obtained the photoluminescence (PL) as a function of the emitted photon energy and objective position shown in Fig. 1. Several PL lines appear at different spatial locations. These lines are due to carriers recombination within single SAQD. Most of the lines appear in pairs. For example, the SAQD located at

10μm emits from three spectral pairs around 1.31, 1.335 and 1.355eV respectively.

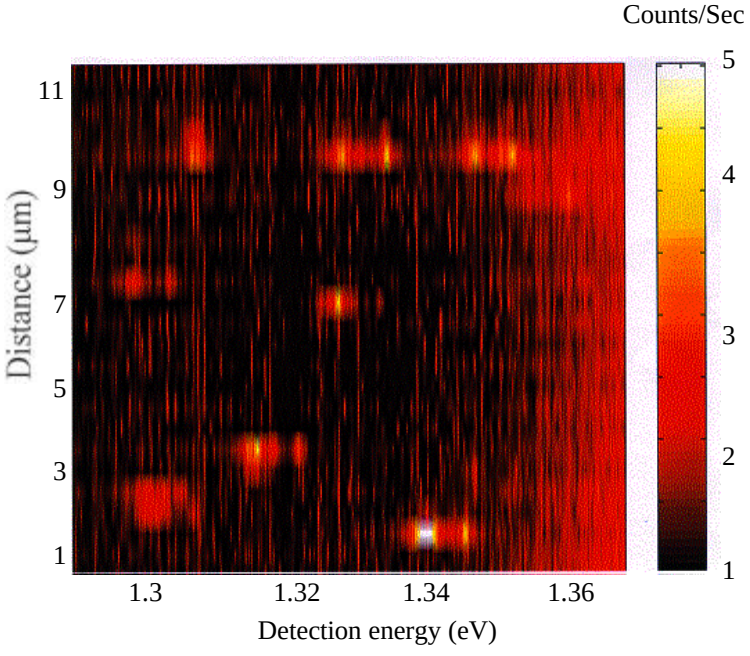

Figure 1. Typical 10K line scan, excited by a 730 nm, 30 μW CW radiation from a Ti:Sapphire laser.

In Fig. 2 we present both CW and pulsed PL spectra from two different SAQDs for various excitation intensities. The observed spectra are very sensitive to the excitation power. The spectrum of both CW and pulsed excitation is composed of two main groups, each of them contains several sharp peaks [2]. The power dependence of the various emission lines is displayed in Fig. 4a. For all lines, the dependence is weaker at higher power levels. But, whereas the weakest line (#3) completely saturates at a power level as low as 20μW, the strongest (#1) shows a square root dependence, with no saturation up to 500μW. The sharp peaks in each transition group are due to the splitting of spin degenerate energy levels. Since each of the single particle quantum confined energy levels can be occupied by up to two particles of opposite spin, a single exciton state should be 4 fold spin degenerate. The degeneracy is higher for larger multi-exciton numbers. We note, however, that the exchange interaction between identical particles reduces this degeneracy [2]. Particularly important is the removal of the 16 fold degeneracy of a multi excitonic state, which contains electrons and holes from two half filled single carrier levels. into three 9,6 and 1 degenerate states, respectively [3]. This splitting is the main reason for the line grouping in the PL spectra [2].

When exciting the SAQD with low power, only the lowest multi exciton states are populated. As the power increases, more excitons occupy the dot, more channels are opened for radiative recombination and as a result more PL lines are resolved. By conducting full many body calculation of the particles that occupy a simple model dot [2], we relate each PL spectrum to the number of excitons that occupy the dot.

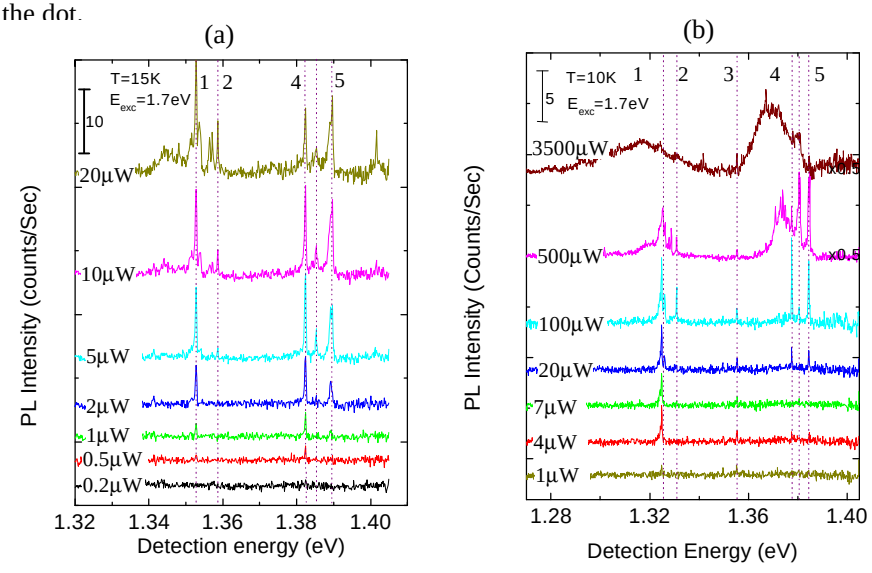

Figure 2. Pulsed (a) and CW (b) PL spectra from a single SAQD for various excitation power level.

We show that the PL spectrum due to recombination of carriers in the first two single particles energy levels is discretely red shifted for increasing number of dot excitons. This trend can be seen in both the CW and pulsed excited PL spectra which are very similar. The main difference between the two is the excitation level which is needed to obtain a similar PL spectra. Whereas in a CW excitation, the number of excitons in the SAQD reaches a steady state, in a pulsed excitation this number is initially larger, it decreases with time and the SAQD is emptied prior to the arrival of the next pulse. Thus, lower average power is required in a pulsed excitation to obtain a similar PL spectrum, and the average PL efficiency is lower in comparison with that obtained with CW excitation.

Starting from a certain n-multi-exciton state in an excited SAQD, we follow its radiative decay to a (n-1)-multi-exciton state with one exciton less. The energies of the emitted photons are equal to the energy difference between the initial **ground** n-exciton level and the final (not necessarily the ground) (n-1)-multi-exciton level. We thus Assume full thermalization of each multi-exciton to its ground level during its radiative lifetime. We likewise follow the decay of

successively smaller multi-excitons, down to the final decay of a single exciton by which the dot returns to its ground state. We then numerically solve a set of coupled rate equations with the radiative oscillator rate of the single exciton, $1/\tau_1$, as the only free parameter (the rest are known from the calculated optical transitions [2]).

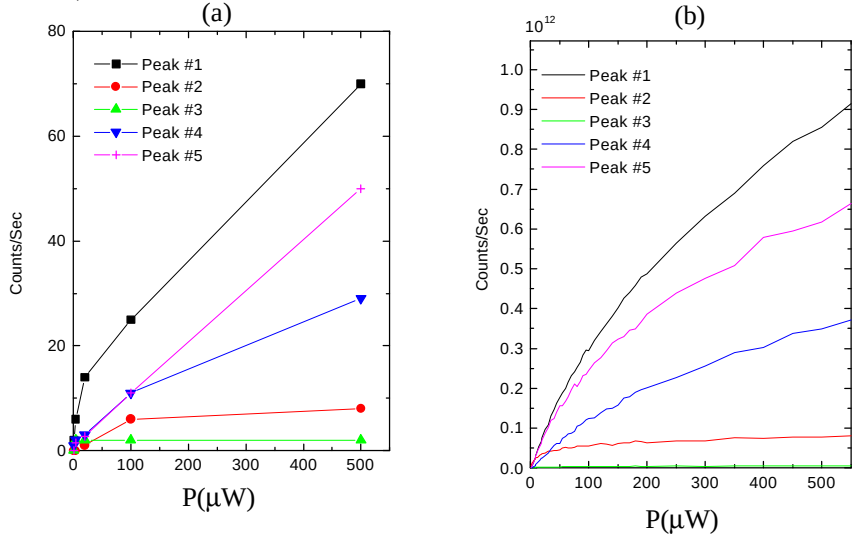

Figure 3. Measured (a) and calculated (b) integrated intensity from the PL lines of Fig. 2, vs. CW excitation power.

Considering up to n=8 multi-excitons states we calculate the power dependence of the dot emission spectrum. For τ_1=1.3 ns, the calculated power dependence shown in Fig. 3b is in good agreement with the experimental data shown in Fig. 3a. These calculations yield τ_2=1.3 ns and τ_n (n>2) approximately 1ns. We therefore conclude that in these SAQDs, photoexcited bound electron hole pairs thermalize into their respective ground multi-exciton state during their radiative lifetime.

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