Time and Space Resolved Spectroscopy of Single Semiconductor Quantum Dots

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We present time-resolved photoluminescence measurements performed on single GaAs/GaAlAs quantum dot structures. Even for an excitation energy below the LO phonon emission threshold very fast PL rise times are observed for all boxes evidencing that even at low excitation power (1-2 electron hole pair in a box) Coulomb scattering plays a major role in the relaxation process. Different recombination dynamics are observed for boxes with various lateral confining potentials: when the lateral confinement is small relaxation occurs within the PL line. On the contrary, when discrete PL lines are present, due to strong lateral confinement, independent recombination of the lines is observed. By increasing the excitation power a particular feature occurs for the box with maximum lateral confinement. Higher energy lines become populated and the spectral weight shifts from the lowest to higher levels. This successive filling of discrete states is interpreted as a consequence of the Pauli exclusion principle assuming the formation of a degenerate Fermi gas of excitons.

I. Introduction

Low dimensionality semiconductor structures seem very attractive both for their fundamental properties and their potential application in optoelectronics. In quantum dots the quantum confinement occurs in all three directions and leads to a discrete density of state. These singularities associated to an increase of the oscillator strengths should give rise to enhanced optical nonlinearities as well as higher performances for laser operation and optical modulators[1-4].

Although techniques of lateral patterning have been greatly improved large inhomogeneities still remain within low dimensional systems arrays. Thus, up to now, most of the experimental results are reported for arrays of OD or lD structures with large fluctuations of sizes. This hampers largely the study of their intrinsic properties.

Space-resolved spectroscopy allows for the investigation of the optical properties of isolated low dimensional systems and has been shown to be succesful in order to observe photoluminescence signals arising from single quantum wires[5] and dots[6]. Under these experimental conditions discrete radiative transitions between quasi zero dimensional states can be measured in semiconductor heterostructures.

In this paper we report on a detailed analysis of the optical properties of single GaAs/GaAlAs quantum dots fabricated by laser-induced local interdiffusion of a quantum well heterostructure. Combining the spatial resolution with time resolution we were able to follow in the time domain the relaxation and recombination properties of these quasi OD systems. As far as these topics are concerned, important questions remain open. It was predicted theoretically that the phonon-assisted relaxation should be considerably slowed down between confined states[7-9]. In quantum dots the relaxation via LA phonon emission should be efficient only between levels with an energy separation lower than a few meV. LO phonon emission (and possibly LO+LA emission) is possible for energies within a few meV from a LO phonon branch[7,9]. Relaxation by Coulomb scattering via an Auger mechanism has also been considered[10]. It is expected to be efficient in the presence of a dense

electron- hole plasma, even for small dots where the phonon scattering rates are low.

In our experiments two parameters are systematically varied, the excitation energy and power. Resonant excitation of the quantum dots is performed above and below the LO phonon emission threshold, in order to evidence the role played by LO phonon scattering in the relaxation process. These measurements are done at low excitation power (between 1-2 and 5-6 excitons per dots). In an other set of measurements we have varied the estimated exciton number in the dot between 1-2 and 200.

The paper is organized as follows. We first describe our samples and experimental set-up. Then we present our experimental findings on the dynamics of our quantum boxes. Finally, an interpretation of the experimental data, based on the appearance of a Fermi gas of excitons for the more confined quantum dot, is given.

II. Samples description and experimental set-up

Our quantum dot stuctures are fabricated by laser-induced thermal interdiffusion of an undoped, 3nm wide, GaAs/$Ga_{0.65}Al_{0.35}As$ quantum well.A dot structure is defined by drawing on the sample surface a square frame of size w with a focused Ar^+ laser. We have studied samples with 2μm, 1μm, 600nm, 500nm, 450nm and 400nm w values. The strongly non-linear temperature dependence of the interdiffusion process allows the realization of steep lateral barriers with about a 35meV height, for both electron and holes. Afterwards, in order to isolate the dot, a 6μ m $\times$6μm square area is interdiffused by scanning the laser beam continuously. Model calculations of the interdiffusion show that near the dot center the lateral confining potential is parabolic[6]. More details on the sample fabrication can be found in Ref. 6 and 11. It is important to stress that the effective size of the lateral confinement is not only given by the geometrical size w but is also modified by the interdiffusion profile. In a general way the lateral size is smaller than the geometrical size.

For large w (2μm, 1μm, 600nm) the lateral confinement is small and a single photoluminescence (PL) line is observed, similar to the one of the 30Å quantum well. By decreasing w, the distance between the lateral barriers decreases and the lateral confinement increases. Discrete PL lines and a blue shift of the fundamental transition, typical of a OD behaviour, appear for w=500nm; for this size the level spacing is moderate, about 3meV. The maximum lateral confinement occurs when the barriers meet at the dot center. For our samples, this situation should be reached for the dot with w=450nm where discrete PL lines with a 10meV splitting are observed. When w is decreased further the Al content at the dot center increases. Thus, the PL line is strongly blue shifted but the lateral barrier height becomes smaller leading to closer PL lines (w= 400nm). For very small values of w the lateral confinement disappears and one deals with an homogeneously alloyed 2D layer.

The samples are mounted on a cold finger in a continuous flow Helium cryostat. Measurements are performed at 7K. The position and the focalisation of the laser on the sample is monitored by a high precision xyz translation stage with an accuracy of about 10 nm. The spot diameter at the sample surface is about 1.5μm (FWHM).

The excitation is provided through a microscope objective by a cw mode-locked Ti: Sapphire laser. It delivers 1.5 ps duration pulses at a 82 Mhz repetition rate in the 685-720 nm wavelength range. Thus, resonant excitation of our structures is made possible. For the photoluminescence measurements the probed area is limited to 1.5 μm by using a pinhole located at an image plane. The PL signal is dispersed by a 32 cm spectrometer and detected by the two-dimensional CCD array of a synchroscan streak camera. The overall spectral and time resolution of our set-up are 2meV and 10ps, respectively.

III. Experimental results

- Dependence on the excitation energy

For all dots we have recorded the PL signal after resonant optical excitation below and above the LO phonon emission threshold (36 meV for GaAs). We were only able to achieve a minimum value of 28meV for the energy mismatch between the fundamental emission and the excitation. For lower values the rejection rate of our set-up was not sufficient and Rayleigh scattering of the excitation pulse hampered a good observation of the PL for short delay times.

We present in Fig. 1a time-resolved spectra obtained for the w=450nm quantum dot structure. For this size the lateral confinement is maximum, discrete PL lines with a 10 meV spacing are observed, and the slowing down of the phonon cascade is expected to be

important. The time dependence of the PL signal is shown for excitation energies 28, 41 and 66 meV apart from the lowest PL line (fundamental transition). In all cases the rise time of the PL signal is fast, of the order of 10ps, close to the time resolution of the experiment. This holds for all the quantum boxes that we have investigated and does not depend on the excitation power.

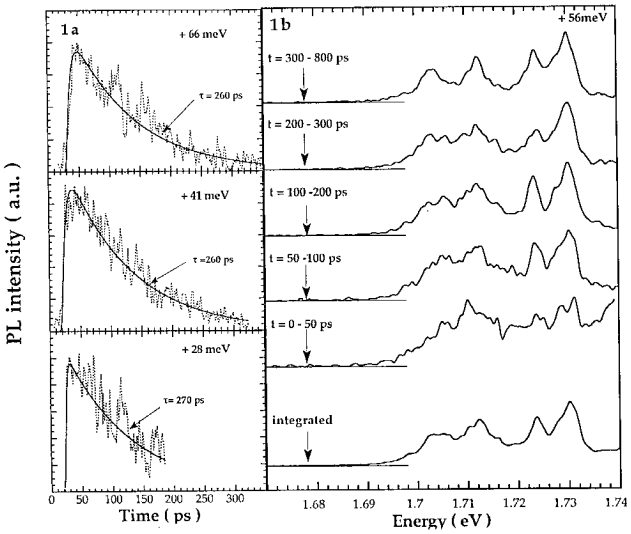

Figure 1. Time decay of the fundamental transition of the w=450 nm quantum dot for three excitation energies above and below the LO phonon emission threshold (1a). PL spectra of the same dot for different time delay windows.

Let us point out that for short delay times the observed PL lines are broadened to an amount which increases with the excitation pulse intensity. This effect can be easily seen in the right part of figure 1 where the temporal evolution of the PL spectrum of the w=450nm box is reported. For this structure the PL lines are well resolved and the effect is clearly evidenced. The four discrete PL lines appear well separated only for large delay times (> 100-200 ps)

Let us now discuss these fast PL rise times. Scattering by LA phonon is too slow to explain the small values of the rise times at least for the w=450nm quantum box. If LO phonon emission were the dominant mechanism for the energy relaxation we would expect a strong dependence of the PL rise time on the excitation energy. A clear slowing down of the relaxation should be measured for excitation energies below the LO phonon emission threshold. We do not observe any dependence of this kind.

As already mentionned, on the contrary, we do measure a spectral broadening of the PL lines at short delay times. The fast measured rise times together with the independence on the excitation energy and the broadening of the lines at short delays indicate that Coulomb scattering contributes predominantly to the energy relaxation process in our quantum boxes. Coulomb scattering can promote a part of the carriers above the optical phonon threshold and allow for relaxation via LO phonon scattering even for small energy differences between the excitation and recombination energy. For quantum dots large relaxation rates are calculated taking into account such an Auger process[10]. Our experimental results show that such a mechanism is important even for very low excitation powers. Low power measurements were done with a 140 nW illumination corresponding to 1-2 excitons per dot.

- Recombination

We have studied samples with geometrical sizes w of 400, 450, 500, 600, 1000, 2000 nm. The insets of Fig. 2 show the time-integrated PL spectra of the boxes with w= 500nm and w=450nm. The 500nm quantum box exhibits a moderate lateral confinement; discrete but not well resolved PL lines are observed with a 3-4 meV spacing. Except for the w=450 nm structure, all the samples show a recombination behaviour which is qualitatively similar to that of the w=500nm one. In all five samples a transfer of intensity from the higher to the lower energy part of the PL spectrum is detected with increasing time. The spectral weight of the PL shifts systematically to the ground state as examplified by the time evolution of the selected spectral windows in the left-side of Fig. 2. The excited states 2 and 3 have recombination times smaller than the one of the fundamental state.

Among the series of samples the w=450nm quantum box structure exhibits the strongest lateral confinement and a strikingly different recombination behaviour is observed. The four spectral windows of the right part of Fig. 2 correspond to the first four energy levels of the box. The decay times measured for the excited states 2, 3 and 4 are similar or even longer than the one of the, fundamental state 1. For this quantum box no transfer of intensity occurs from higher to lower energy.

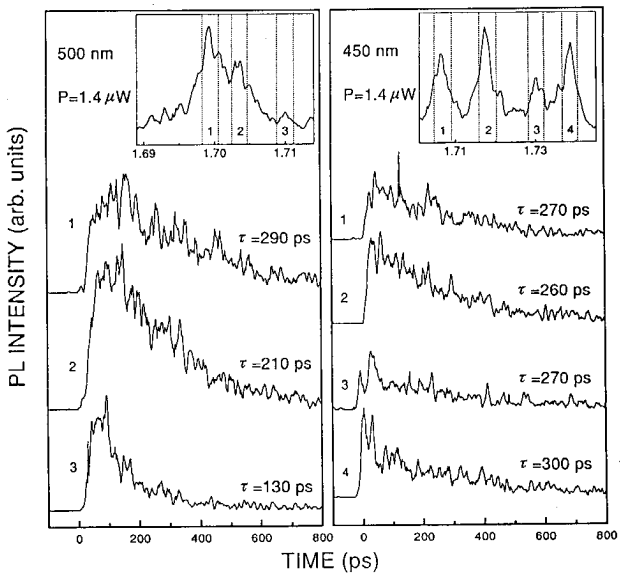

Figure 2. Time dependence of the PL intensity for different spectral windows in the w = 500 nm (left part) and w = 450 nm quantum dot structures. The insets show the time-integrated PL spectra.

- Dependence on the excitation power

Fig. 3 shows time-integrated PL spectra of the w=450 nm quantum box for various excitation powers. They are recorded without changing the lateral position and the focalisation of the laser spot. For a 140 nW excitation power we estimate that we create 1.7 exciton in the dot. Two PL lines separated by about 10meV are observable, the fundamental one being more intense. For 450 nW, these two peaks are still present but now the excited state gives a stronger PL signal. For increasing excitation powers two strong, additional lines appear at higher energy and the two low energy peaks saturate. The spectral weight of the PL shifts systematically to the excited states.

On the contrary the other samples seem to obey to a classical energy distribution. For increasing excitation powers the PL spectrum broadens; recombination arising from high energy states can be observed but the spectral weight of the PL is always centered on the fundamental transition. Let us finally note that we do not observe any noticeable shift of the PL lines with increasing excitation power. For instance the lowest two lines of Fig. 3 shifts by less than 1 meV in the power range investigated. The higher energy levels do not seem to shift. We have no experimental evidence of correlation effects such as band-gap renormalisation.

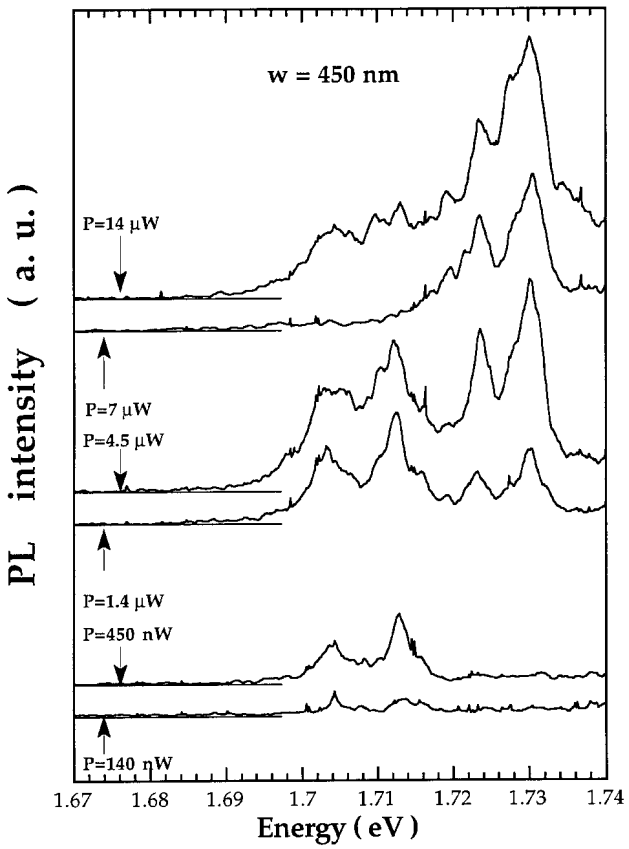

Figure 3. Excitation power dependence of the PL spectrum of the w = 450 nm quantum dot structure.

IV. Interpretation and discussion

The strong and unexpected power dependence of the PL spectra of the w=450 nm quantum dot is interpreted in terms of a successive filling of excited states. In that particular case the excitons in the quantum dot behave like fermions. In fact, this not so surprising. For the 450nm dot the spatial extension of the fundamental state is estimated to be 30-40 nm and has the same order of magnitude that the 3D exciton Bohr radius in GaAs (15nm). Thus, a kind of Mott transition is expected to occur with a moderate number of excitons in the dot. In consequence, for quantum dots exhibiting a noticeable lateral confinement, it does not exist a real low density regime corresponding to an exciton gas of Bose statistics. The key feature responsible for the particular behaviour of the 450 nm dot is the stronger lateral confinement i. e. the large energy separation between the distinct PL peaks. The Fermi energy of the exciton gas is defined by filling one after the other the discrete levels of the box. Due to the large spacing

of these levels existing in the 450nm box the Fermi energy exceeds rapidly the thermal energy of the exciton gas (the fourth excited level is located 30meV above the fundamental one). A degenerate Fermi distribution is easily reached; this is not achieved in the other boxes where the level spacing is much smaller. In other words in these structures the density of states is still too high to achieve a degenerate gas.

Let us consider now the power dependence of the 450 nm dot PL spectrum. The lowest states first acquire a sizeable occupation. The level degeneracy is higher for excited levels[12], thus the lower peaks saturate while the upper ones increase quasi linearly with the excitation power. For high excitation powers the upper lines with a higher population number become dominant. On the contrary for the other boxes the relative weight of the different recombination lines qualitatively follow a classical statistics.

The formation of a degenerate Fermi gas of excitons also explains the different recombination behaviours exhibited by the 450 nm quantum dot. With increasing lateral confinement, relaxation via LA phonon scattering is expected to be considerably hampered[7,8]. This may explain why higher energy transitions are observed even for low excitation powers. Nevertheless we have shown that Coulomb scattering is the predominant mechanism for the energy relaxation in our samples, thus we rather consider that as soon as a sizeable part of the states is occupied, the relaxation is hindered due to the Pauli exclusion principle. This is only possible in the 450nm dot where the large level spacing (i. e. the low density of state) allows the formation of a degenerate Fermi gas of excitons.

V. Conclusion

We have performed time-resolved spectroscopy on single quantum dots containing only 1-2 excitons. Coulomb scattering appears to be the predominant mechanism for energy relaxation in these dots: A density dependent broadening of the PL lines is systematically observed at short time delays, the PL rise times are always fast and do not depend on the excitation energy. We do not observe the predicted slowing down of phonon emission with increasing lateral confinement.

For the quantum dot with maximum lateral confinement we were able to populate significantly excited states more than 30meV above the fundamental one. Moreover, all populated states seem to recombine almost independently which is not the case for less confined quantum dots. These experimental findings are well interpreted assuming the formation of a degenerate gas of excitons. Our experimental results seem to evidence the fermionic character of the quantum dot excitons.

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