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CHARGING OF SELF-ASSEMBLED INAS SINGLE QUANTUM DOTS BY PURE OPTICAL MEANS

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ABSTRACT

We have studied self-assembled InAs single quantum dots (QDs) using a conventional μ -PL setup to detect PL from an individual QD. The excitonic features, neutral and negatively charged, have been identified in these QDs. For certain experimental conditions, what concerns the laser excitation energy, the laser power and the sample temperature, drastic differences in the line spectra are monitored. These differences are interpreted in terms of charging/discharging of the quantum dots. This phenomenon is predominantly determined by the excitation energy of the laser and is proposed as an effective optical method to control the charging of the QDs.

1. INTRODUCTION

Self-assembled quantum dots (QDs) can be considered as an artificial atoms because QDs, in contrast to the structures of higher dimensionalities, effectively confine electrons (e) and holes (h) on the nanometer length scale in all three directions. This results in zero dimensional density of states as well as in the increased role of Coulomb interactions in QDs [1]. The latter together with the very restricted number (typically two) of particles which could be accommodated at the ground state of the QD, makes the role of multiparticle effects of great importance both from the point of view of fundamental research and for variety of applications. Indeed, multiparticle states, consisting of a large number of e and h (equal or non-equal) in QDs, which have been studied theoretically [1-5] and experimentally [6-13], determine the performance of a number of optoelectronic (electronic) devices, for example QD lasers and single-electron transistors, which in turn are considerably affected by the charge, stored in the QD [14].

This highlights the important role of multiparticle complexes with non-equal number of e and h , and hence stimulates the study of charged exciton complexes, which in the simplest case consist of one exciton and one additional charge carrier (i.e. e or h). Also more complex charged excitons with several charge carriers of the same sign can be formed [2]. For example, strong dependence of the QD emission spectra on the number of extra electrons (up to 20) was predicted in [2], where the role of the band gap renormalization as well as the shifts of the spectral lines in the charged exciton complexes due to the exchange energy were discussed in detail. Experimental evidence for the formation of few electron states in QDs was revealed in capacitance experiments [7, 11] and studies of interband transmission [9,10]. The formation of negatively charged exciton complexes was demonstrated in PL experiments [6, 8]. However, optical studies [8-10] were dealing with a large ensemble of dots, so the

results inevitably include the effect of inhomogeneous broadening which prevents accurate measurements and hence detailed analysis of optical properties of the QD. That is why the studies of charged exciton phenomena in the luminescence of single dots are highly desirable.

Up to date two approaches have been used to study charged exciton complexes in single QDs. One method is to use specially designed samples [12] containing an n-doped layer with contacts. The number of electrons in the QD can then be tuned by an applied external voltage. This gives a possibility to control the average number of excess electrons in the dot. However, the applied voltage causes a shift of the entire spectrum and deforms the wavefunctions, which eventually leads to changes in the interaction energies. The other method is to use samples where the QDs are initially filled with a random number of electrons from the background-doping [13]. The main drawback of this method is that the initial number of excess electrons depends on the arbitrary number of impurity atoms in the close vicinity of the investigated QD and hence hardly controllable.

2. SAMPLES

The samples studied were grown by Molecular Beam Epitaxy in Stranski-Krastanow mode on the semiinsulating GaAs (100) substrate. On the top of the GaAs barrier layer, the QDs are formed from a 1.7 InAs monolayers deposition. Transmission electron microscopy studies of analogously grown samples revealed that uncapped original dots are lens (hemispherical) shaped with a typical lateral size of 35 nm and a height of 4.5 nm. For the μ -PL characterization, certain sample regions of very low dot density was chose, where the interdot spacing was typically of the order 10 μ m, well exceeding the resolving power of the μ -PL setup used.

3. EXPERIMENTAL SETUP

The QDs were studied by means of a conventional diffraction-limited μ -PL setup (See Fig. 1). To excite the QDs, a cw beam of Ti-Sp laser tunable between 700 to 900 nm was used, which could be focused on the sample surface down to 1.5 - 2 μ m spot in diameter by a microscope objective through a thin optical window of the continuous flow cryostat (See Fig. 1). The luminescence signal was collected by the same objective and dispersed by a single-grating 0.45 m monochromator combined with a LN₂ cooled Si-CCD camera, which gives the spectral resolution in the region of the studied PL of 0.15 meV.

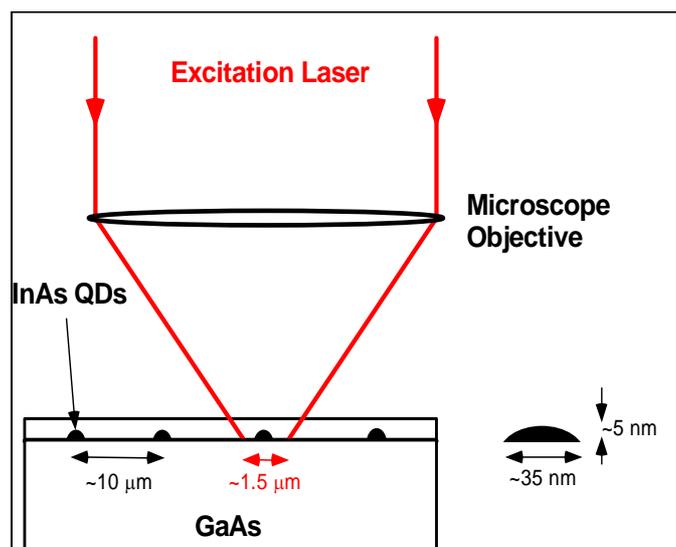


Fig. 1 Laser beam focused down to a small spot in the μ -PL setup.

To find the particular QD to study, we a laser beam was scanned across the sample surface. Once the desired QD was found, special marks (grids) were fabricated on the sample surface around the QD with the laser beam of very high power density. This allowed us to estimate the average distance between the adjacent QDs to be around 10 μm in the studied QD structure. To control the exact position of the laser spot on the sample surface, the image of the interesting sample region was projected by a video camera, which made it easy to find the desired QD by using the fabricated marks. In addition, this arrangement allowed us to effectively correct the position of the sample if it was moved due to the thermal drift. It should be noted that with the method to locate the exact QD position with the use of the described grids one can avoid some undesirable consequences which take place for other methods.

3. OPTICAL CHARGING BY ABOVE BANDGAP EXCITATION IN μPL

Fig. 2 shows $\mu\text{-PL}$ spectra of an individual QD taken at a temperature of $T = 4\text{ K}$ and two different exciting photon energies above the GaAs bandgap.

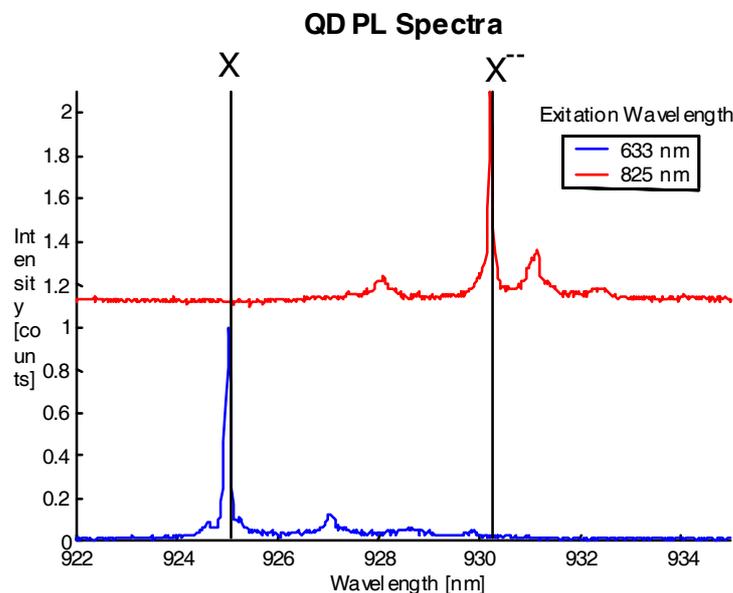


Fig.2. μPL spectra of an individual QD at $T=4\text{ K}$, for two different pump-photon energies.

At low P_{ex} in spectra taken at an excitation wavelength of 633 nm (Fig. 2), a single line is revealed, which is labelled as X. This line is very narrow with its full width at half maximum of 0.15 meV, which is believed to be limited by our spectral resolution. The absence of other lines implies that the PL signal registered originates from the emission of a single QD. At another excitation wavelength of 820 nm, the $\mu\text{-PL}$ spectrum clearly shows the appearance of novel lines, with the line marked as X^- dominating. To understand the dramatic changes in PL initiated by the tuning of the excitation energy, we note first, that the difference occurs at lowest excitation intensity levels, so this allows us to exclude from the explanation possible influence of multiexcitonic complexes related to the occupation of QDs with more than one exciton ($e\text{-}h$ pair). To understand the dramatic changes in PL initiated by the tuning of the excitation energy, we note first, that the difference occurs at lowest excitation intensity levels, so this allows us to exclude from the explanation possible influence of multiexcitonic complexes related to the occupation of QDs with more than one exciton ($e\text{-}h$ pair). Second, the appearance of X^- and X^- lines can not be explained as impurity related transitions because these would appear at any excitation conditions. Third, both excitation energies used are above the GaAs barrier and hence, the observed changes in PL spectra can not be understood in terms of the different influence of the below and above barrier excitations on the QD luminescence.

On the basis of these theoretical predictions [8, 9], the lines marked as X^- and X^{--} are ascribed as the charged exciton complexes with one and two additional electrons, respectively. Consequently, some characteristic energies, such as the binding energy of the single charged exciton $E_{(X^-)^b}$ and the exchange energy $E_{sp}^{(ex)}$ (between s and p electrons with parallel spins, as described below) can be deduced from the experiment: $E_{(X^-)^b} = E_{(X)} - E_{(X^-)} = 3.1$ meV and $E_{sp}^{(ex)} = E_{(X^-)} - E_{(X^{--})} = 4.7$ meV, where $E_{(X)}$, $E_{(X^-)}$ and $E_{(X^{--})}$ correspond to the spectral positions of the X, X^- and X^{--} lines respectively.

At an excitation wave length of 825 nm (Fig.2), the QD is accordingly populated with additional electrons, while in the case of pumping with an excitation wave length of 633 nm, there are no extra electrons captured to the dot. These differences are attributed to the different excess energies of electrons at these two excitations. To further elucidate the role of electron excess energy, the dependence of the PL spectra on the pump-photon energies $h\nu$ was investigated. Fig.3 shows the evolution of the PL peak amplitude of X and X^- lines as a function of $h\nu$. The oscillating behavior of both X and X^- is clearly seen in Fig. 3. It is important to note that the amplitudes of X and X^- behave in a reversed way, i.e. when X gains its maximum value, X^- acquires its minimum amplitude and vice versa. Accordingly, every time the QD is populated with extra electrons, the creation probability (and hence the PL amplitude) of the X^- line increases, while the corresponding probability for X decreases. The possible difference for the two $h\nu$'s used in case of Fig. 2 is explained in terms of different excess energies (with respect to the band gap E_g) of electrons, ΔE_e , and holes, ΔE_h , photoexcited in the GaAs barrier. A carrier can release its excess energy via the emission of optical or acoustic phonons. If ΔE_e (ΔE_h) is larger than the optical phonon energy (for example a longitudinal optical phonon $\hbar\omega_{LO} = 36$ meV in GaAs, then the particle will effectively release its energy via the cascade emission of LO phonons. These processes are very fast (of the order of a few ps) and hence the "cooling" time for the particle, for example e , will be determined by the residual excess energy $\Delta E_e^r = \Delta E_e - n\hbar\omega_{LO}$, where $n = 0,1,2$. The latter can be released only via the emission of acoustic phonons which takes much longer time. Hence the higher the ΔE_e^r (ΔE_h^r) is, the longer time is required for e (h) to cool down, form the exciton and recombine. This eventually leads to the conclusion that particles with larger ΔE^r are able to move a longer distance before the exciton is formed, or in other words, they possess higher effective diffusivities

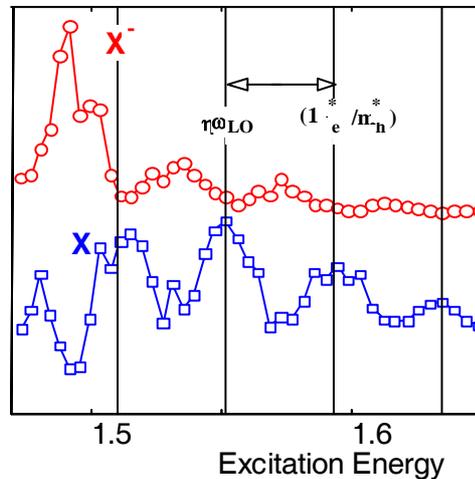


Fig.3. The PL intensities of X and X^- lines as a function of the excitation energies.

This picture should repeat itself whenever the electron acquires the same residual excess energy. The latter, in turn, takes place every time pump-photon changes its energy by the value of $\Delta h\nu = \hbar\omega_{LO} \times (1 + m_e^*/m_h^*)$ which follows from the simple picture of band-to-band excitation in direct band gap semiconductors in the approximation of parabolic valence and conduction bands. Taking the electron and heavy hole effective masses in GaAs as $m_e^* = 0.067m_0$ and $m_h^* = 0.45m_0$ and $\hbar\omega_{LO} = 36$ meV, we obtain $\Delta h\nu = 41.4$ meV, which is in very good agreement with the period of the oscillation observed in Fig. 3.

5. OPTICAL CHARGING BY BELOW BANDGAP EXCITATION IN μ PL.

Next, the effect of selective excitation below the GaAs bandgap will be investigated. Two μ PL-spectra of a single QD are shown in Fig. 4, taken at two adjacent excitation energies, namely below and above the crucial energy of 1.483 eV, situated below the barrier band gap ($E_g^{\text{GaAs}} = 1.519$ eV), but above the emission of the WL (~ 1.45 eV). The two spectra are very different: The neutral exciton (X) and the negatively charged exciton (X^-) are essentially the only lines present in the low-excitation energy spectrum, while X and X^- have vanished completely to the favor of the double negatively charged exciton (X^{2-}) in the high energy spectrum. The abrupt change from a strongly negatively charge-state to an almost neutral state is clearly demonstrated in the upper panel of Fig. 5, in which the intensities of X and X^- are recorded as a function of the excitation energy, revealing a step-like behavior at 1.483 eV.

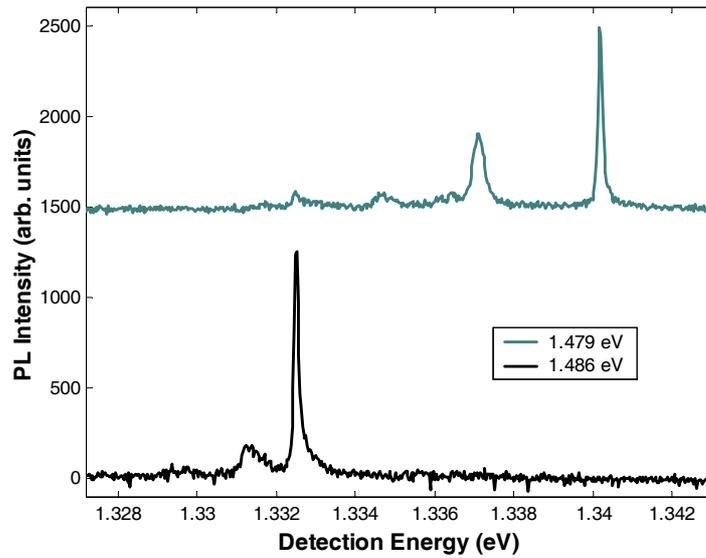


Fig.4 . μ PL spectra of an individual QD at $T=4$ K, for two different pump-photon energies.

All QDs studied exhibit a similar abruptly change their charge-state. However, the energy at which the change occurs depends on the QD position; the transition energy is increased for QDs situated in regions of less InAs deposition, corresponding to regions with a thinner WL.

The lower panel of Fig. 5 exhibits the behavior of another QD positioned in a region of lower InAs deposition. The step-like behavior remains but it is blue shifted by about 9 meV compared with the dot in the upper spectrum. The observed dependence of the step energy on the QD position strongly suggests that an energy level associated with the WL is involved in the charging process of the QDs.

The step in Fig. 5 is explained in terms of an impurity induced photo generation of excess electrons above a threshold energy, $h\nu_{th}$. Upon laser excitation with $h\nu_{ex} > h\nu_{th}$, an equal number of e^- 's and h^+ 's, will be created. However, in addition some free electrons will be photo-generated in the surrounding barriers as a result of the light absorption by the ionized acceptor atoms, which are present in the barrier as residual dopants. Then, the $h\nu_{th}$ has to be equal to the $E_g^{\text{GaAs}} - E_a$, where E_a is the acceptor binding energy. In fact, the $h\nu_{th}$ values, obtained for high P_{ex} for the two groups of QD's, would imply E_a to be equal to 36 (26) meV for the first (second) QD's group, respectively. These values are similar to the Si and C acceptor binding energies of 34 and 25.5 meV, respectively. The extra electrons created in the GaAs barriers, are expected to be effectively captured into the WL (and subsequently into the QD) due to their very efficient transport in GaAs accompanied by the absence of free holes in the barriers at $h\nu_{ex} < E_g^{\text{GaAs}}$.

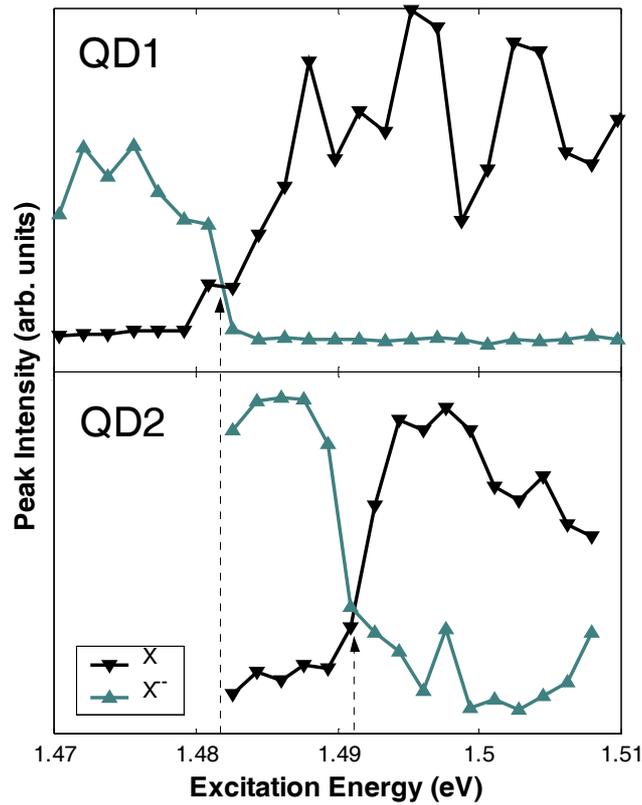


Fig. 5. μ PL spectra of an individual QD at $T=4$ K, for two different pump-photon energies.

Accordingly, in order to populate the QD with a charged exciton configuration, absorption of at least two photons is needed. One of them excites an exciton inside the WL (see Fig. 6) and the second should excite an electron in the GaAs barrier (Fig. 6). This results in an expected superlinear dependence ($\sim P_{\text{ex}}^m$) of the PL amplitude of the charged exciton lines on P_{ex} .

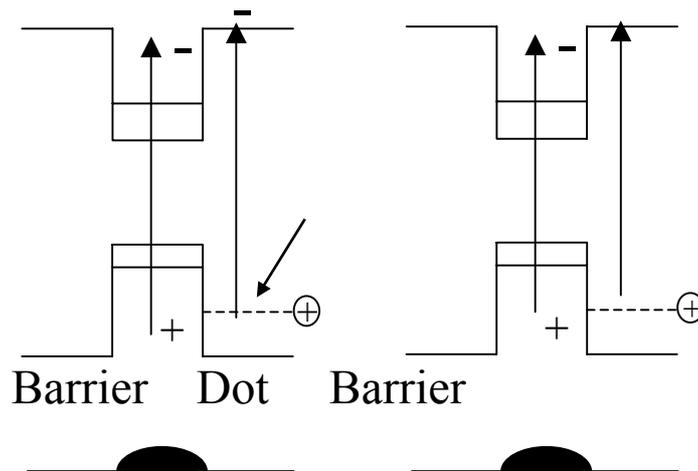


Fig. 6. An illustration of the transitions involved in the formation of the neutral and the charged excitons in the dot by performing excitation below and above, respectively, the threshold energy corresponding to the acceptor to the conduction band in the surrounding GaAs.

6. Summary

In summary, we have demonstrated that photoexcited or injected carriers in the barrier will be captured into the QD from a rather large surrounding area, typically a few μm . Important parameters affecting this capture process are e.g. the effective mass of the diffusing particles, the temperature and the excess energy of the created carriers relatively the band edge of the barrier material. The dependence on the excess energy can be studied by scanning the excitation energy above the barrier in $\mu\text{-PL}$ measurements. The intensities of the charged and neutral exciton, respectively, in InAs/GaAs QDs exhibit an oscillatory and anti-correlating dependence. This behavior is explained in terms of different diffusivity of the electrons and holes. The excess energy of the photo-generated carriers can be released by emission of fast LO phonons. If all the excess energy is released by emission of an integer number of LO phonons, the carrier will end up without kinetic energy at the band edge to form neutral excitons. Otherwise, the remaining kinetic energy will be transferred into carrier diffusion. The capturing dot will primarily trap the high diffusivity electron due to the smaller mass, resulting in the formation of negatively charged excitons. This method will accordingly allow us to create and study neutral and charged exciton complexes in single QDs by pure optical means.

Furthermore, we have shown that the dot can be seen as an extremely sensitive probe for trapping of additional carriers, when the excitation energy is tuned above or below a well-defined threshold energy. In our case the threshold energy is associated with an acceptor level in the vicinity of the dot. However, the charge state of the QD could be used to monitor also other defect states in the neighborhood of the dot, under the assumption that a tunable laser is accessible in the relevant energy region. This means that the quantum dot could be utilized as an extremely sensitive charge probe for defect states in the vicinity of the dot.

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