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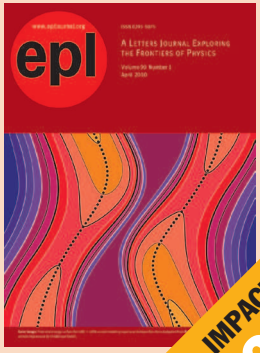

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Recombination dynamics of excitons and exciton complexes in single quantum dots

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Abstract – We have studied the recombination dynamics of excitons and excitonic complexes confined in single GaAs quantum dots, embedded in a type-II GaAs/AlAs bilayer, formed at unintended growth imperfections. The small density of defects leads to the spatial isolation of the quantum dots, allowing to address individual specimens without any further sample processing. Any influence of carrier diffusion on the recombination dynamics is avoided by using quasi-resonant excitation, below the quantum dot barrier. Under these excitation conditions, the recombination occurs within a 2 nanosecond time window since relaxation takes place only inside the quantum dot. At low powers, the photoluminescence spectra are dominated by very sharp lines attributed to the exciton and the bi-exciton/charged-exciton transitions, while at large powers it is possible to observe the emission from higher-order exciton complexes. We have found a retardation of the emission increasing the pump power and interpreted it as an evidence for a sequential decay of the different excitonic species.

Introduction. – The lack of complete perfection in the crystal growth of two-dimensional (2D) semiconductors when taken as an asset has greatly contributed to the investigation of quantum dots (QD) [1]. Among representative examples of such structures are laterally confined 2D objects, which result from fluctuations in the width of the 2D layer (quantum well) [2,3]. When probed with micro-photoluminescence (PL) experiments, these QDs raise sharp emission lines , which emerge from the broad macro-PL spectra due to a large collection of localized 2D excitons. The QDs due to potential fluctuations are weakly confined in the lateral direction but strongly in the growth direction. Their formation has been reported in different (preferentially narrow) quantum well structures [4,5].

Previous studies [6,7] show that growth imperfections in a type-II GaAs/AlAs bilayer may also lead to a formation of QDs that are very different from those resulting from well width fluctuations. The density of such QDs is very low and they give rise to sharp emission lines in a very wide energy range. It was proposed that the origin of this kind of QDs is the inclusion of direct band-gap GaAlAs, surrounded by the original type-II GaAs/AlAs bilayer, and that they are strongly confined systems in all three dimensions. The emission spectra of these QDs, in the limit of low excitation power, present several distinct lines, assigned to single neutral- (X), bi- (XX) or charged- (X^*) exciton recombination. More generally and typically for strongly confined systems, with few atomic-like shells, the PL spectra are found to be significantly affected by the excitation power [8–13]. Time-resolved (tr) spectroscopy appears to be a relevant method to study such systems [14–16].

Here we investigate the relaxation and emission dynamics of excitons and different exciton complexes confined in single QDs that appear on a type-II GaAs/AlAs bilayer. We tune the excitation energy below the GaAlAs barrier in order to photo-generate carriers only inside the QD. A sequential emission is clearly seen in the time-resolved spectra: the recombination from the higher-order exciton complexes occurs before the bi-exciton recombination,

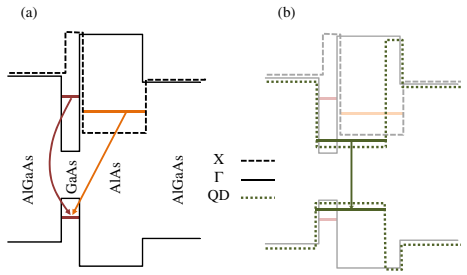

Fig. 1: (Colour on-line) (a) Band alignment of the bilayer structure, showing the allowed optical transitions from the Γ- and the X-points in the conduction band. (b) Band alignment resulting from the formation of QDs, *i.e.* Ga-rich islands, at growth imperfections. The faded lines depict the bilayer band alignment, for comparison.

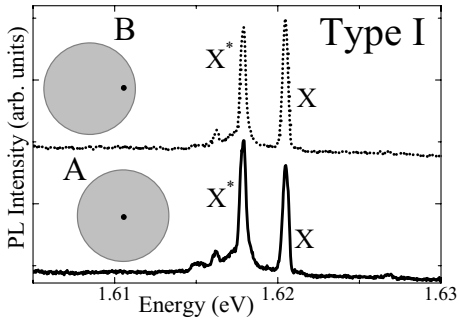

Fig. 2: Time-integrated PL spectra obtained from two different positions of the excitation spot over a Type-I dot (positions A and B). The gray circle/black dot schematically represent the relative positions of the excitation spot/QD. The spectra are obtained under pulsed excitation (32 μW) and for a temperature of 12 K. The X and X^* transitions are observed.

which takes place prior to the single exciton recombination. Eventually, at very long time delays, the tr-PL spectra resemble those obtained under continuous wave excitation conditions at low excitation powers.

Experimental details. – The sample under study consists of a type-II GaAs/AlAs double quantum well structure [6,17,18]. The appropriate choice of both GaAs (2.4 nm) and AlAs (10 nm) layer thicknesses allows to observe not only the direct recombination (Γ-Γ) in the GaAs quantum well but also the indirect recombination (X-Γ) in the AlAs/GaAs bilayer (fig. 1(a)), which becomes optically allowed due to a weakening of the translational invariance [19,20]. We assume that during the sample growth, unintended gallium-rich islands form at layer imperfections. In these islands, the GaAs/AlAs bilayer is replaced by GaAs/$Ga_{1-x}Al_xAs$ clusters ($x < 0.33$), from which the recombination appears in the form of very sharp peaks. These peaks are attributed to dot-like emission. The changes in the band alignment associated with the QD structure are schematically displayed in fig. 1(b). This dot-like emission is observed in a wide spectral range (from $\sim$ 1.56 to $\sim$ 1.7 eV), reflecting the large fluctuations in the QDs composition and/or thickness, the latter being limited by the total bilayer width (*i.e.*, 12.4 nm). Kelvin force microscopy [21] measurements reveal an average QD size of tens of nanometers and a QD density as low as $10^6\,cm^{-2}$ [19]. Such a low QD density allows to excite individual QDs without using metallic masks.

The optical excitation of single QDs is performed using 2 ps long pulses obtained from a $Ti:Al_2O_3$ laser, tuned to 740 nm and spectrally narrowed with an interference filter, which removes the residual super fluorescence of the laser. The excitation wavelength, below the $Ga_{1-x}Al_xAs$ energy gap, is selected in order to quasi-resonantly create electron-hole pairs inside the QDs, pumping into the high-energy exciton complexes states. This makes it possible to almost completely neutralize the influence of carrier diffusion and trapping on the recombination dynamics, that otherwise would be present when pumping into the $Ga_{1-x}Al_xAs$ surrounding layer. The laser is focused to a 1.5 μm diameter spot using a 50× microscope objective mounted on 3 motorized translation stages (14 nm spatial precision), which is also used to collect the QD-PL. The sample is inside a cold finger cryostat where its temperature is kept at 12 K. To reject the residual laser light, the emitted PL is filtered with an edge filter (cut at λ = 752 nm) placed at the entrance slit of an imaging spectrograph (spectral resolution 100 μeV). The time-integrated PL is detected with a standard CCD coupled to one of the exits of the spectrograph while the tr-PL is obtained with a streak camera coupled to the second exit of the spectrograph, working on the photon counting mode and acquiring for several tens of minutes (*i.e.*, integrating over several billions of recombination events). The overall time resolution is $\sim$ 50 ps.

Results and discussion. – The first step in our experiments is the identification of the different transitions observed in the PL spectra. To do so, we have performed standard micro-PL measurements and we have found two different kinds of dots in the sample. One sort of dots in which the coupling to the neighboring charged states is very large (Type-I dots) and their PL spectrum shows both, X and charged-exciton X^* recombination lines. In the other kind of dots (Type-II dots) the coupling to the neighboring charged states is negligible and the emission is predominantly due to the recombination of neutral-X complexes (X recombination at low power and XX at higher powers).

Let us first describe the situation observed in Type-I dots, whose characteristic time-integrated spectra are shown in fig. 2. The fingerprint of these dots is the presence of two emission lines even in the limit of the lowest excitation powers, which are attributed to X and X^* recombination. To determine whether the charge bound with the exciton is an electron or a hole is a difficult task for III/V-based heterostructures. We can argue that X^* is a negatively charged exciton from an inspection of the GaAlAs barrier material and the GaAs buffer layer

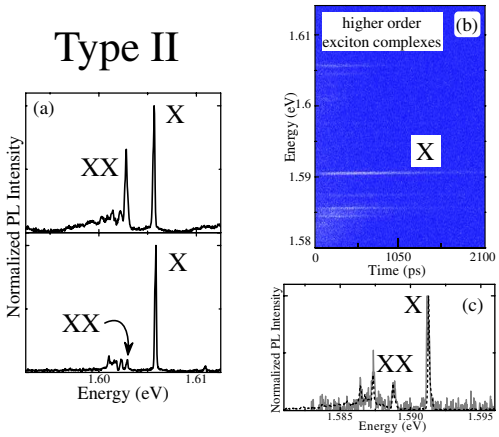

Fig. 3: (Colour on-line) (a) Time-integrated emission of Type-II dots obtained after pulsed excitation for 32/220 μW (bottom/top panel, respectively). The spectra display the X and the XX line. (b) Streak camera image displaying the time-resolved QD-PL from both the X transition and the recombination from higher-order exciton complexes (excitation power 32 μW). (c) Normalized time-integrated emission (black dashed line) and time-resolved spectrum (grey line) measured for a delay time of 600 ps and a pump power of 270 μW.

PL spectra, which shows an overall n-type remote doping of our structure. However, this is not a robust enough argument to discard the binding of a hole. In any case, the actual sign of the charge has a small impact on the general conclusions drawn from our results, so we will refer to the additional line observed in the QD-PL spectra as the recombination of a charged exciton, without specifying the sign of the charge. Characteristically these dots are pretty fragile in the sense that the actual intensity ratio of X and X^* is very sensitive to the alignment of the laser spot with respect to the dot location (see fig. 2). At low power levels, the ratio of X and X^* emission is unaffected by small changes on the excitation power. However, the X^* emission is enhanced if the laser spot is centered on the dot (position A). A small displacement of the laser spot, either in the horizontal or in the vertical direction (position B), results in a reduction of the X^* emission when compared to that of the X recombination. Using below-barrier excitation we expect to inject the carriers into the dot in the form of electron-hole pairs and therefore postulate that the simultaneous observation of X and X^* emission is due to the fluctuating charge state of the dot (*e.g.*, the oscillatory tunneling of an electron between the dot and the surrounding defects). Changes in the intensity ratio of X and X^* show therefore that the dynamics of the fluctuating charge critically depends on the excitation conditions, which likely influence the apparent electrostatic potential around the dot [22]. The experimental results summarized in the present manuscript have all been obtained centering the excitation spot on the different QDs.

The results of the optical characterization of Type-II dots are remarkably different (fig. 3(a)). In these QDs, at low pump power (bottom graph), the PL spectrum is dominated by a single emission line attributed to the X recombination. These dots are much more robust as compared to Type-I dots. Their spectrum does also evolve as a function of excitation power but showing a more conventional behavior: the progressive appearance of the bi-exciton (XX) and eventually higher-order multi-excitons, which give raise to the emission below the X line as well as at higher energies, far above the X line (not shown). In the limit of high excitation power, the multi-excitonic emission regroups into a sequence of rather broad but still well separated bands, which are associated with the zero-dimensional energy levels (resembling atomic shells) of the dot.

The tr-PL spectra, obtained on a different Type-II dot, whose characteristic transitions appear at slightly different energies, are very similar to the time-integrated ones, as can be seen on figs. 3(b), (c): the exciton emission is observed, together with the recombination from bi-excitons and higher-order excitonic complexes, which are shown now on either energy side of the X emission line. The streak camera image displayed in panel (b) shows that after quasi-resonant pulsed excitation the majority of the X recombination processes take place within a 2 ns time window, while for out of resonance excitation (above the QD barriers), the characteristic recombination times are governed by indirect-exciton diffusion and lie in the μs range [23–26]. The emission due to higher-order excitons (below 1.584 eV and at $\sim$1.605 eV) is relevant at short times after the laser excitation (< 500 ps). At later times, the emission spectrum consolidates in the dominant X recombination. The direct correlation between the time-integrated and the tr-PL is clearly seen on fig. 3(c), where the tr-PL obtained 600 ps after the arrival of the excitation pulse (gray line) overlaps almost perfectly with the time-integrated one (dashed line).

Let us now discuss the excitation power influence on the X recombination dynamics. The main results obtained on Type-I dots are summarized in fig. 4. The two top panels ((a) and (c)) display the raw, time-resolved images for different pump powers. Two very narrow lines appear on the images: the X at 1.598 eV, the X^* at 1.5951 eV, yielding an X^* binding energy of $\sim$ 2.9 meV, and other transitions that are tentatively attributed to a third charge-related state or a bi-exciton at 1.5945 eV. We have found that the X^* binding energy ranges from 2.2 meV to $\sim$ 3 meV [19], increasing with the dot size, in a similar way as that reported in quantum wells [27]. It is observed that the recombination of X and X^* start simultaneously and shortly after the arrival of the excitation pulse, since the X^* recombination cannot generate any X and vice versa [28]. Also we note that the X^* recombination is slightly faster than that of the X [29]. At higher powers it is possible to see the hot bulk GaAs PL at $t = 0$ ps, appearing as a horizontal white stripe (fig. 4(c)).

For Type-I dots, the X and X^* emission are the dominant radiative recombination channels at any time

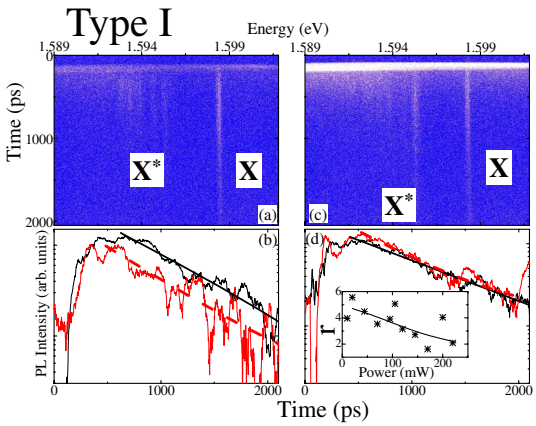

Fig. 4: (Colour on-line) The top two graphs display raw streak camera images for different pump powers: (a) 105 μW and (b) 220 μW. The bottom two graphs show the corresponding semi-logarithmic plot of the time evolution traces of the X (black) and the X^* emission (red). These time evolution traces are obtained after a 50 adjacent points smoothing of the raw traces. The inset in panel (d) corresponds to an estimation of the ratio between the time the charge stays outside (τ_{out}) and inside (τ_{in}) of the QD, $r = \tau_{out}/\tau_{in}$. The line is a guide to the eye.

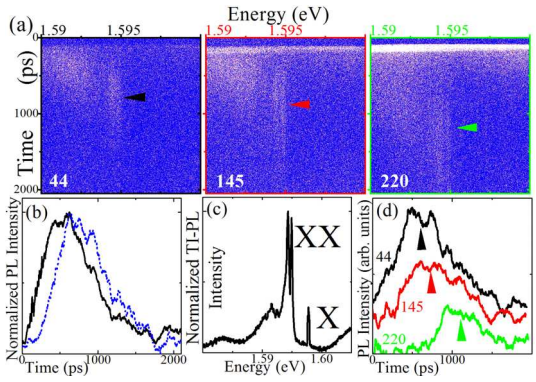

Fig. 5: (Colour on-line) (a) Streak camera images for different pump powers (44, 145 and 220 μW, respectively) displaying the time evolution of the XX recombination. The arrows mark the time position of the emission maximum. (b) 50 adjacent points smoothed time evolution traces corresponding to the higher-order excitonic complexes/XX recombination (solid line/blue dotted line) for a 44 μW pump power. (c) Normalized time-integrated PL obtained for 270 μW. (d) XX emission time evolution traces for the same pump powers as on (a) after a 50 adjacent points smoothing.

delay and laser power investigated. Their time evolution traces are displayed on the bottom panels of fig. 4 (X/X^* in black/red). The QD emission intensity is so small that in order to improve the signal to noise ratio we have performed a 50 adjacent points smoothing of the time evolution traces. To gather some insight on the recombination dynamics we have analyzed the time evolution traces, extracting two characteristic times: the decay time (τ_d), fitting the decaying part of the time evolution traces with a mono-exponential decay function, and the time to reach the maximum emission intensity (t_{max}), directly obtained from the time evolution traces. We observe an increase of τ_d for both X and X^* with pump power, increasing from 675/455 ps to 880/600 ps for X/X^*, respectively. The observed retardation of the X and X^* decay with the laser power might arise from a local modification of the potential landscape around the dot, which possibly implies changes in the efficiency of the non-radiative recombination channels. These times are considerably smaller than those recently reported for InAs/GaAs QDs coupled in a similar way to the electrostatic environment [22]. The difference in the decay times is due to the different (quasi-resonant) excitation conditions considered here.

Taking advantage of the fact that X^* and X are independent recombination channels [28] we have estimated the ratio between the time the charge stays out (τ_{out}) and in (τ_{in}) the QD, $r = \tau_{out}/\tau_{in}$: when the charge is out/in, the emission originates from X/X^* so we have calculated r integrating the area underneath the time evolution traces of X and X^*. The dependence of r on pump power is summarized in the inset of fig. 4(d). At low powers, the charge stays longer outside the QD ($r \sim 5$), but as the power is increased, the two times become more alike ($r \sim 2$) and the two transitions have comparable amplitude. Such a balance between the X and the X^* recombinations has been observed recently [30] and justified in terms of an increase in the average charge of the QD with excitation power.

A similar retardation of the dynamics of the X emission, but of different origin, is observed in Type-II dots. These dots can be more easily populated with many photo-excited electron-hole pairs. Figure 5(a) shows the raw streak camera images for increasing pump power. At short times ($t < 500/1000$ ps for 44/220 μW) the PL is dominated by the broad emission from higher-order excitonic complexes, appearing between 1.59 and 1.5925 eV. The presence of a large number of carriers, both electrons and holes, favors the formation of bi-excitons. The XX recombination, at 1.5948 eV (blue dotted line, fig. 5(b)), starts once the dot is emptied enough and the emission from the higher-order excitonic complexes (solid line, fig. 5(b)) starts to decay. The X emission cannot be seen on the streak camera images not only because of its faint intensity but also because it will occur at very long times, much longer than the 2 ns time window of the detector. Yet it is clearly observed on the time-integrated PL (fig. 5(c)), at $\sim$ 1.598 eV, together with the XX state and the multi-excitonic emission. A clear retardation is observed on the XX dynamics increasing the pump power (fig. 5(d)), directly reflected in t_{max} [31], which continuously increases from ~ 590 ps to ~ 1020 ps increasing the pump power from 44 to 220 μW. Here, as in the case of Type-I dots, it is the multi-excitonic radiative cascade

the mechanism behind this retardation of the dynamics [22,32,33]. Regarding the decay dynamics, fig. 5(d) reveals that the XX time evolution trace develops a non-exponential decay behavior with increasing pump power. The weak PL intensity of the bi-exciton recombination hinders the extraction of reliable information about τ_d, making the analysis of τ_d's power dependence a very delicate task.

Conclusions. – We have time-resolved the emission of a single QD after quasi-resonant excitation. This suppresses the influence of diffusion and trapping of carriers on the recombination dynamics. The large 3D confinement of our dots allows us to observe multi-excitonic complexes, such as the charged-exciton X^* and the bi-exciton XX, together with higher-order excitonic complexes. The increase of the carrier density confined inside the QD (increase of pump power) is directly reflected in the PL spectrum and its dynamics: at very low powers, only the emission from X^* (in Type-I dots) and X (in Type-I and Type-II dots) are observed; at higher powers, the emission from higher-order excitonic complexes occurs at early times, followed by the multi-excitonic complexes emission and finally the X recombination. The retardation of the dynamics caused by the recombination cascade is evidenced by a rise of both the decay time (τ_d) and the time delay for maximum emission intensity (t_{max}), which almost double their values with an increase of the pump power by about two orders of magnitude.

* * *

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