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Exciton radiative lifetime controlled by the lateral confinement energy in a single quantum dot

J. Hours, P. Senellart,* E. Peter, A. Cavanna, and J. Bloch
CNRS-Laboratoire de Photonique et Nanostructures, Route de Nozay, 91460 Marcoussis, France
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We report on time-resolved measurements on single GaAs quantum dots formed at the interface fluctuation of a GaAs/AlGaAs quantum well. We measure exciton radiative lifetimes as short as 100 ps, demonstrating that monolayer fluctuation quantum dots have larger oscillator strength than any other *III-V* or *II-VI* semiconductor quantum dots. Studying various single quantum dots, we demonstrate that the oscillator strength of a quantum dot is controlled by its lateral confinement energy.

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The recent intensive study of semiconductor quantum dots (QDs) has opened up the possibility of achieving optical quantum devices based on single quantum dots emission.[1–4] Recently, understanding the role of Coulomb interaction in the multicarrier emission from a single QD (Ref. 5) has led to a demonstration of a single photon source in solid-state physics.[2] To go further and demonstrate quantum operations based on QDs, different options are investigated. One option largely investigated[6] is to achieve all optical quantum gates. This requires that the decoherence time is limited by the radiative lifetime of the QD. Yet, most of the time, reported decoherence times are much shorter than the measured radiative lifetimes. A solution to this problem has been found in the use of the spontaneous emission enhancement occurring when the QD is placed in an optical microcavity.[3,6,7] An alternative way could be to use QDs with shorter radiative lifetimes. Another option investigated to realize quantum operations is the possibility to couple different QDs through the interaction with an electromagnetic field.[8] To do so, the strong-coupling regime for a QD in a microcavity,[9–11] where the radiative emission is reversible, has to be achieved. In the two options described above, QDs with short radiative lifetimes or, in other words, large oscillator strengths, are strongly needed.

QDs formed at an interface fluctuation of a quantum well are very attractive for this purpose.[12] It has been theoretically predicted that they should present a much larger oscillator strength than other *III-V* QDs.[13] Indeed, recent single QD absorption measurements have enabled the measurement of an oscillator strength around $f=45-180$.[14] Besides, Andreani and co-workers[13] have calculated that the oscillator strength of these QDs should be controlled by the lateral size of the fluctuation QD.

We have recently demonstrated that monolayer fluctuation QDs can deliver a single photon emission with a repetition time smaller than 200 ps.[15] In this paper, we report on time-resolved measurements on single monolayer fluctuation QDs. By studying the emission dynamics as a function of excitation power, we evidence the radiative cascade between the biexciton emission and the exciton emission. We measure radiative lifetimes as short as 100 ps for the exciton state and 60 ps for the biexciton state. This exciton radiative lifetime corresponds to an oscillator strength of $f=75$ as compared to $f=9$ for InAs QDs. By studying various single QDs, we evidence lifetimes ranging from 100 to 230 ps. We correlate these measurements to single QD photoluminescence excitation (PLE) measurements that give access to the exciton confinement energy, and as a result, to the lateral size of the QD.[16] We experimentally show that there is a direct correlation between the QD lateral confinement energy (and thus its lateral size) and its oscillator strength; the oscillator strength goes to a minimum and increases significantly for both the small and large QD radius.

The sample under study consists of a nominally 10 monolayer GaAs quantum well embedded in $Al_{0.33}Ga_{0.67}As$ barriers. The detailed description of the sample growth and its optical characterization can be found in Ref. 15. To isolate single QDs and increase the collected signal, microdisk structures have been chemically etched.[17] All measurements are performed at a nominal sample temperature of 5 K. Microphotoluminescence (μPL) measurements are performed in the far field using a microscope objective. We excite the sample with a Ti:sapphire laser delivering 3 ps pulses every 12 ns. The excitation is nonresonant with an energy around 1.73 eV. The sample emission is spectrally dispersed using a 32-cm spectrometer and temporally analyzed with a streak camera system. For the measurements discussed in the following, the spectral (temporal) resolution is 0.25 meV (20 ps).

Figure 1 presents three streak camera images obtained for three excitation powers: $P_0=7$ W/cm^2, $2P_0$, and $5P_0$. On each image, the horizontal direction corresponds to energy, the vertical direction to time. The intensity of the emission is represented using a color scale. On top of each image, the time-integrated spectrum is plotted. The high-energy broad emission is attributed to the recombination of excitons in the two-dimensional (2D) quantum well (QW) corresponding to the upper energy monolayer region. Notice that localized exciton states are observable as discrete lines in this broad emission. This localization arises from interface roughness. On the low-energy side of the spectrum, we observe the emission from a spectrally well isolated single quantum dot. At low excitation power P_0, the emission essentially arises from the recombination of a single electron-hole pair inside the QD, at the exciton (X) energy of 1.6977 eV. On the left side of Fig. 1, the temporal dependence of the X emission is plotted. The X emission rise time is very short (around 30 ps), which evidences an efficient capture process inside the QD. For long-time delays (around 500 ps), the X emis-

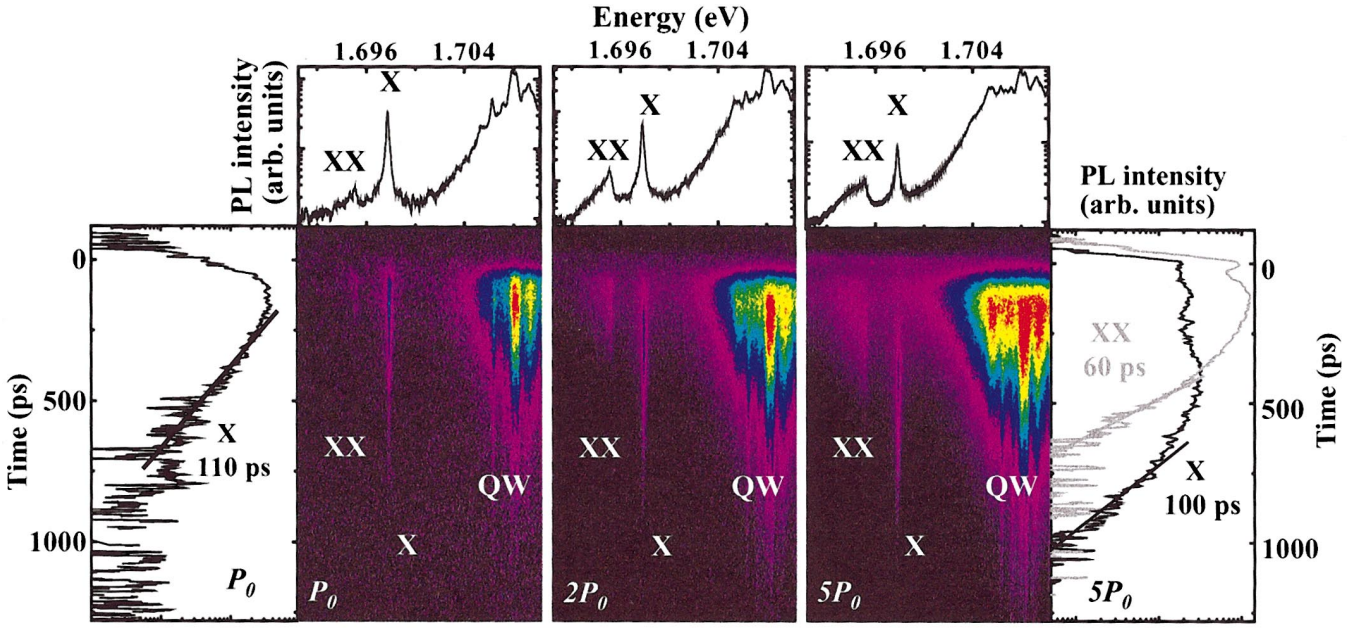

FIG. 1. (Color) Streak camera images measured for three excitation powers: $P_0=7$ W/cm^2, $2P_0$, and $5P_0$. The horizontal scale corresponds to energy, the vertical scale to time. The top of each image: time integrated spectrum (logarithmic scale). Left (respectively, right) side of the images: spectrally integrated emission as a function of time for *X* (black) and for *XX* (gray), for P_0 (respectively, $5P_0$). The lines are a linear fit to the emission decay for long time delay.

sion decays monoexponentially with a decay time of 110 ps. For higher excitation powers ($2P_0$ and $5P_0$), the biexciton (*XX*) emission, signature of the presence of two excitons in the QD, appears at an energy around 1.695 eV. The *XX* emission rises shortly after the excitation pulse, whereas the *X* emission is delayed. This appears clearly in the time dependence of the photoluminescence signal shown in the right part of Fig. 1 for $5P_0$. This time-resolved measurement gives a direct evidence of the radiative cascade, usually evidenced in many QD systems by time-correlation measurements.[5,18] For large delays, both *X* and *XX* emissions decay monoexponentially with decay times of $t_X=100$ ps and $t_{XX}=60$ ps. These decay times are independent on the excitation power.

We note that both the *X* and *XX* decay times are shorter than the 160 ps QW emission decay time. For the two highest excitation powers, the *XX* line presents a peculiar line shape: a broad emission sideband develops on the low energy side of the line. Considering the overall emission of the sample on the streak camera images, we understand the origin of the *XX* low-energy sideband. At very short time delays and for high excitation powers, the *XX* recombines while other electron-hole pairs are still present in the surrounding QW. The Coulomb interaction of these electron-hole pairs with the *XX* inside the QD results in a redshift of the *XX* emission.[19] The larger the time delay is, the smaller the population inside the QW is, and the less the *XX* emission is redshifted. Finally, when the *X* is emitted, at long time delays, the QW is empty. We show elsewhere that this low-energy sideband on the *XX* line is the signature of fast emitting QDs.

In Fig. 2, we have plotted the spectrally and temporally integrated emission from both the *X* and *XX* emission line. For the *XX* line, the integrated intensity includes the low-energy sideband. At low excitation power, the *XX* line presents the characteristic quadratic power dependence while the *X* line grows linearly. We model the power dependance of both lines taking into account the fact that an electron-hole pair can be recaptured by the QD after the *XX* emission. This recapture is particularly important in these QDs because, as already mentioned above, the *XX* lifetime is shorter than the lifetime of excitons in the 2D QW. The calculated intensities, shown in solid lines on Fig. 2, very well reproduce the measured intensities.

Finally, we have compared the absolute signal collected at the exciton saturation from these QDs to the signal collected from InAs QDs, in equivalent microstructures. The signal at saturation is 10 times larger with GaAs QDs than for InAs QDs, which is consistent with a 10-times-smaller radiative lifetime. We can thus conclude that the decay time we measure is mainly radiative and that non radiative mechanisms can be neglected.

Up to now, few direct measurements of single QD dynam-

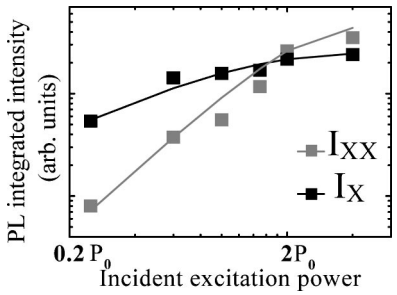

FIG. 2. Squares: spectrally and temporally integrated intensity of the *X* line (black) and of the *XX* line (gray) as a function of the excitation power. Full lines: calculated *X* and *XX* intensities.

ics have been reported.[20–23] The radiative lifetime of InAs QD excitons ranges from 400 ps to 1.9 ns. The oscillator strength can simply be deduced from the radiative lifetime through[13]

$$f = \frac{1}{t_X}\frac{3\pi\epsilon_0}{n}\frac{2mc^3}{e^2\omega_0^2}.$$

ω_0 is the optical transition frequency, n the refractive index and m is the free-electron mass. Thus the measured lifetime for InAs QDs corresponds to an oscillator strength from $f=6$ to $f=25$.[5,22] In *II-VI* QDs, radiative lifetimes around 300 ps (i.e., $f\approx 13$) have been reported.[20] The oscillator strength corresponding to the X radiative lifetime we measure is $f=75$. Our finding is consistent with the single monolayer fluctuation QD absorption measurement reported in Ref. 14, and show that GaAs QDs present the largest oscillator strength. Recently we have inserted such QDs in a micrometer-sized microdisk and observed the strong coupling regime with a Rabi splitting of 400 μeV, twice larger than the individual linewidths.[11] This large Rabi splitting as compared to values measured with self-organized In-based QDs (Refs. 9 and 10) is another signature of the record oscillator strength of GaAs monolayer fluctuation QDs.

We now study the radiative lifetime of various QDs. Among all QDs studied, the X lifetime ranges from 100 to 230 ps. To understand the origin of these variations, we have performed PLE measurements on each single QD. Figures 3(a) and 3(b) show typical PLE spectra obtained for two QDs (QD_α and QD_β), presenting exciton radiative lifetimes of $t_\alpha=100$ ps and $t_\beta=150$ ps. These PLE spectra exhibit a clear absorption step corresponding to the absorption of the QW 2D continuum. The energy difference ΔE we measure between the X emission line and this 2D absorption edge is the lateral confinement energy of the exciton within the QD. We have $\Delta E_\alpha=12$ meV for QD_α and $\Delta E_\beta=3.7$ meV for QD_β. In Fig. 3(c), we have reported the measured radiative lifetime as a function of ΔE. The vertical and horizontal bars represent the experimental error in our measurements (typically 1 meV for the confinement energy and 20 ps for the radiative lifetime). For very small confinement energies, the radiative lifetime is very short (around 100 ps). It then increases and reaches a maximum around a confinement energy of 4–5 meV. For larger confinement energies, the radiative lifetime decreases continuously, and reaches 100 ps again.

The exciton confinement energy inside the QD is directly related to the lateral size of the monolayer fluctuation QD. The smaller the radius of the QD is, the smaller ΔE is. To be more quantitative, we model the confinement of the exciton in these QDs. Since here the electron-hole Coulombic attraction is stronger than the lateral confinement potential, we consider a quasi-two-dimensional exciton whose center of mass is weakly localized at the QW interface fluctuations. In our model, we consider a Gaussian potential in the QW plane: $V(R_{xy})=-V_b e^{-R_{xy}^2/2a^2}$, where R_{xy} is the exciton center of the mass position, V_b is the height of the potential, and $\sqrt{2}a$ is the monolayer fluctuation QD lateral radius. V_b is determined by macrophotoluminescence measurements as in

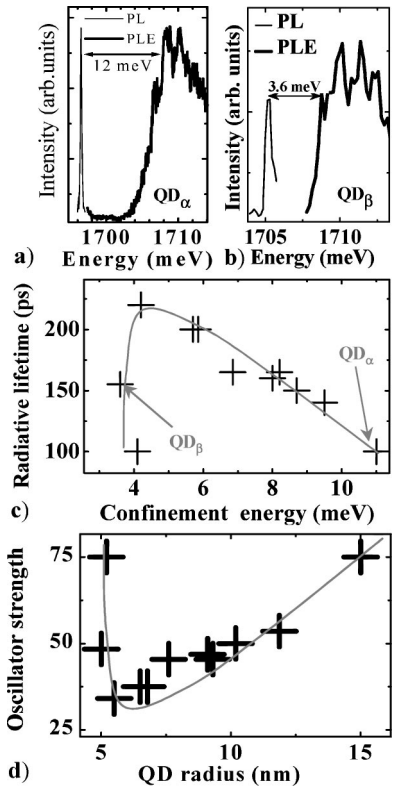

FIG. 3. (a),(b) PL (thin line) and PLE (thick line) spectra for two QDs. (c) Symbols: radiative decay time as a function of the exciton confinement energy inside the QD. (d) Symbols: oscillator strength as a function of the QD lateral radius. (c),(d) The line is a guide to the eyes.

Ref. 16. We have $V_b=17$ meV. The localized exciton wave function is $\psi_X(r_e,r_h)\propto e^{-R_{xy}^2/2\xi^2}e^{-\rho/\lambda}$, where ρ describes the electron-hole relative motion and λ is the exciton Bohr radius. ξ is a variational parameter describing the lateral extension of the center of mass movement. We finally solve the variational problem for ξ and relate the confinement energy of the exciton center of mass to the monolayer fluctuation QD lateral radius $\sqrt{2}a$.

In Fig. 3(d), we plot the oscillator strength of the QDs as a function of their radius. The oscillator strength goes to a minimum for a QD radius around 6 nm and increases for both the smaller and larger radius. This observation is in very good agreement with the dependence predicted by Andreani and his co-workers.[13] This dependence can be understood considering that the oscillator strength is proportional to the area occupied by the center of mass. On one hand, for large radius QDs, the center of mass is localized in a quasi-two-dimensional trap. Therefore, its wave function occupies an area increasing with the QD radius. The oscillator strength is of the order of the two dimensional QW oscillator strength (per area units), multiplied by the area of the monolayer fluctuation QD. Recently Langbein and co-workers have reported a similar decrease of the radiative lifetime with confinement energy using four-wave mixing measurements on several ensembles of InGaAs QDs.[24] On the other hand, for small QD radii, the center of mass is weakly localized in the QD. The wave function of the electron and hole state ex-

pands outside the defect, inside the barrier. The smaller the QD is, the more these wave functions expand, and the larger the oscillator strength gets.

Finally, notice that the dependence of the oscillator strength reported in Fig. 3(d) is not in perfect quantitative agreement with the theoretical predictions of Ref. 13. This can be explained by several reasons: the model we have used to deduce the quantum dot radius from ΔE significantly depends on the precise shape of the defect (cylindrical, rectangular, etc.) Moreover, the X wave function we use factorizes the relative motion and the center of mass motion; this approximation is good for large QD radii, but not so good for very small radii.

To conclude, by performing time-resolved measurements on single GaAs monolayer fluctuation QDs, we have evidenced that the exciton radiative lifetime can be as short as 100 ps. By studying several QDs presenting various radiative lifetimes, we show that the oscillator strength of the QD is directly controlled by the confinement energy of the exciton inside the QD, and consequently by the lateral size of the QD. The typical oscillator strength $f=75$ that we measure for large QD radius is one order of magnitude larger than the oscillator strength of self-assembled InAs QDs, which make them promising for cavity quantum electrodynamics experiments in the strong-coupling regime.

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*Electronic address: pascale.senellart@lpn.cnrs.fr

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