Nanotechnology **17** (2006) 1831–1834 doi:10.1088/0957-4484/17/8/004

Neutral and charged multi-exciton complexes in single InAs quantum dots grown on InP(001)

N Chauvin[1], E Tranvouez[1], G Bremond[1], G Guillot[1], C Bru-Chevallier[1], E Dupuy[2], P Regreny[2], M Gendry[2] and G Patriarche[3]

[1] LPM, UMR CNRS 5511, INSA de Lyon, 7 avenue Jean Capelle, 69621 Villeurbanne Cedex, France
[2] LEOM, UMR CNRS 5512, Ecole Centrale de Lyon, 69134 Ecully Cedex, France
[3] LPN, UPR CNRS 20, Route de Nozay, 91460 Marcoussis, France

E-mail: nicolas.chauvin@insa-lyon.fr

Received 4 October 2005, in final form 5 December 2005
Published 15 March 2006
Online at stacks.iop.org/Nano/17/1831

Abstract
Low density InAs/InP(001) quantum dots, formed by the ripening of self-organized quantum sticks, are subjected to optical spectroscopy by photoluminescence and micro-photoluminescence. The presence of neutral multi-exciton states in single quantum dots is observed up to the 4X state. The large energy spacing between the s- and p-shells (90 meV) reveals the strong spatial confinement of carriers in such InAs/InP quantum dots. Micro-photoluminescence studies show that some dots are charged which allows us to measure the exchange energies in the InAs/InP quantum dots.

1. Introduction

Single InAs/InP(001) quantum dots (QDs) are promising structures for the realization of various devices emitting in the 1.55 μm spectral range. For instance, these QDs could be used in single photon emitters at optical telecommunication wavelengths [1], or in quantum computing processing devices. The design and realization of nanosources containing an isolated single QD in a submicrometre area needs intense research efforts in both domains: in molecular beam epitaxy (MBE) in order to better control the shape and density of InAs QDs grown on InP(001) substrates and in optical spectroscopy in order to get an accurate knowledge of the discrete energy states of such QDs in the 1.3–1.55 μm spectral range. The determination of such QDs energy states are of prime importance for single photon emission application.

In this paper, we present an optical spectroscopic study of single InAs QDs grown by solid-source MBE on InP(001). Specific growth conditions are chosen in order to favour the ripening of the elongated quantum sticks (QSs) which are usually obtained in this material system [2–4]. Photoluminescence (PL), micro-photoluminescence (μ-PL) and polarized μ-PL are performed on single InAs/InP quantum dots emitting around the 1.55 μm wavelength. This optical spectroscopic study allows the observation of neutral and charged multiexcitons, as well as the determination of the biexciton binding energy, the energy spacing between the s- and p-shells and the exchange energies in InAs/InP QDs.

2. Experiment

The InAs QD sample is grown by solid-source MBE on a semi-insulating InP(001) substrate. One plane of InAs QSs is grown with the deposition of 1.6 ML of InAs and growth parameters which reduce the stick size dispersion (growth temperature at 510 °C with an As2 pressure of 2×10^{-6} Torr and a growth rate of about 0.22 monolayer (ML) s^{-1}) [2]. Then, the InAs QDs are obtained by the ripening of uncapped QSs during the cooling of the sample to about 430 °C under arsenic overpressure. The ripening process was followed by atomic force microscopy (AFM) on samples cooled at different temperatures [5]. It appears that ripening is not complete at 430 °C and that some residual sticks can still be present in the sample studied. Finally, for PL measurements, the InAs QDs are capped with a 17 nm-thick InP layer.

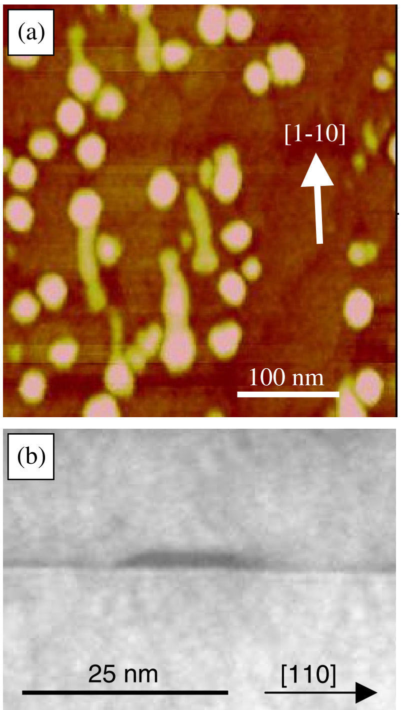

Figure 1. (a) AFM and (b) [110] cross-sectional TEM dark field images of the InAs/InP QDs.

(This figure is in colour only in the electronic version)

The excitation for the PL experimental setup is performed using the 514.5 nm line of an argon-ion laser. The single QD spectra are obtained by μ-PL measurements. The μ-PL experiments are realized through nano-apertures made in a 30 nm-thick gold mask, with an average diameter of 100 nm. The local excitation for the μ-PL experiment is provided by a He–Ne laser at 632.8 nm, through a microscope objective which focuses the excitation power on a 2 μm-diameter surface. The sample is mounted in a He-flow cryostat and maintained at about 8 K. The ultimate spectral resolution of our experimental setup is 200 μeV.

3. Results and discussion

Figure 1(a) shows an atomic force microscopy (AFM) image of the uncapped coalesced InAs QDs. Most of them are spherical with a mean diameter and height of 30 and 4 nm, respectively, and with a density of 1.3×10^{10} dots cm^{-2}. Some others, such as the InAs QSs, are due to incomplete ripening and remain elongated in the $[1\bar{1}0]$ direction. The size of the circular dots have to be compared with QSs obtained previously on InP(001) substrates, whose mean width and length are 22 and 75 nm respectively [2]. The [110] cross-sectional TEM image of the studied capped sample (figure 1(b)) shows the typical shape and size of the obtained InAs QDs after InP capping. The lateral size and height of the capped InAs QDs are 21 ± 5 nm and 2.2 ± 0.5 nm, respectively. The difference in the sizes deduced from the AFM image is principally explained by the size modification due to As/P exchange during the growth of the InP capping layer and also to an overestimation of the lateral size due to the AFM tip convolution. The PL spectra

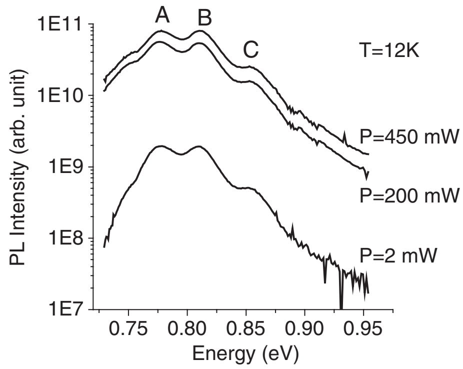

Figure 2. Macro-PL spectrum as a function of the excitation power density.

of the capped InAs QDs recorded at 12 K (figure 2) show several peaks. The evolution of the spectra as a function of the excitation power density indicates that these peaks are related to different sets of QDs due to monolayer (ML) height fluctuations.

Micro-photoluminescence of single QDs is performed through different nano-apertures. Figure 3(a) clearly shows the ground state (s-shell) and the first excited state (p-shell) of one of these single QDs as a function of the excitation power at a temperature of 8 K. The energy spacing between the two shells is 90 meV. Such a value is much larger than the value (around 25 meV) previously measured in InAs/InP QSs whose large dimension is as high as 75 nm [6]. But it is in agreement with a theoretical study performed on lens-shaped InAs/InP quantum dots, where an energy spacing of 100 meV is calculated for a lens-shaped QD of 17.5 nm diameter [7].

A close-up of the s-shell is shown in figure 3(b). At low power density, only one emission line labelled X is observed at 838.2 meV. The presence of this single μ-PL peak at low power excitation confirms the absence of any bottleneck phonon effect in the studied single QD. Thus, we can assume that the μ-PL peak originates from the recombination of electron–hole pairs in their lowest energy states, and that peak X originates from the recombination of the exciton ground state (GS) in the QD. When the power density is increased, a second peak (labelled 2X) appears at 834.2 meV. The integrated intensity of this peak increases quadratically with the excitation power and is associated with the biexciton recombination. The biexciton binding energy is defined by $B_{XX} = 2E(X) - E(XX)$, where $E(X)$ and $E(XX)$ are the exciton and biexciton ground states, respectively. This value is $B_{XX} = 4$ meV in the QD under study. For an excitation power of 30 μW, one single peak arises in the p-shell at 930 meV (peak labelled 3X in figure 3 (a) and is associated with the transition of the triexciton GS (initial state) to the biexciton GS (final state): 3X(GS) $\rightarrow$ 2X(GS).

When the power density is further increased, a second peak labelled 4X emerges below the 3X peak and then dominates the spectrum: this peak is associated with the transition from the 4X GS to the 3X GS. The energy shift between the transitions 4X(GS) $\rightarrow$ 3X(GS) and 3X(GS) $\rightarrow$ 2X(GS) (5.9 meV) is greater than the biexciton binding energy (4 meV). Thus, adding a second electron to the p-shell results in a more stable state than adding a second electron to the s-

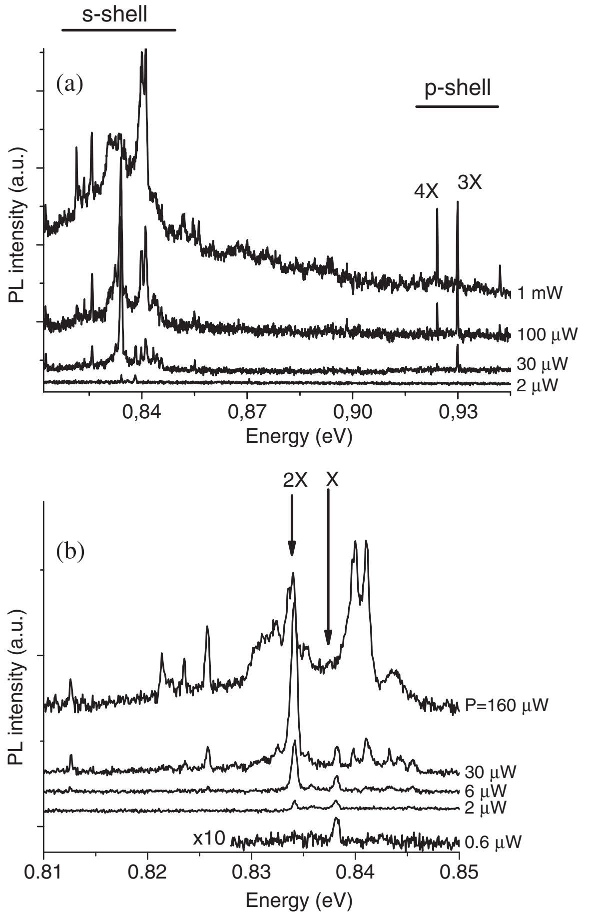

Figure 3. μ-PL spectra of a single QD as a function of the excitation power density: (a) observation s-shell and p-shell. (b) Zoom on the s-shell.

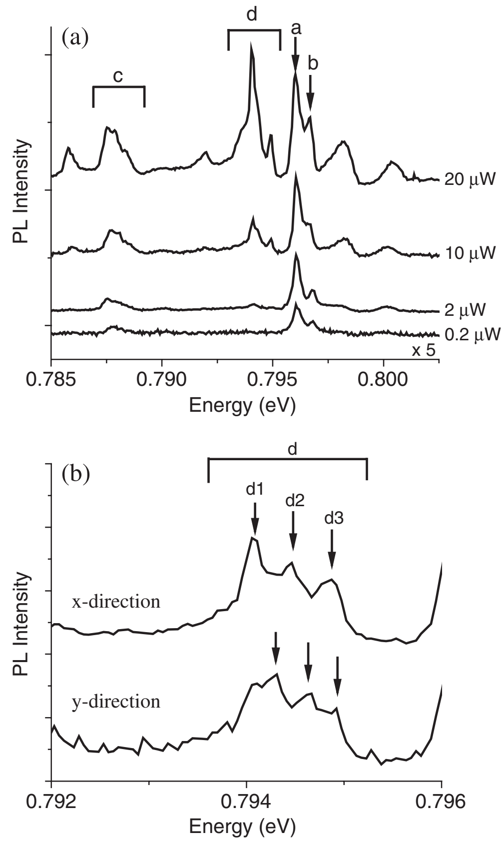

Figure 4. (a) μ-PL spectra of a double charged single QD as a function of the excitation power density. (b) zoom on the μ-PL spectra of the $2X^{2-}$ state measured along cross-polarized directions.

shell. At a higher excitation density other peaks appear in the s and p-shell due to higher multi-exciton complexes.

Similar results are also observed in other single QDs in this sample. The biexciton binding energy ranges between 4 and 5.5 meV with a mean value of 4.5 meV. The large biexciton binding energy (4 meV) and the large separation between the ground and first excited states (about 90 meV) imply a strong spatial confinement. These values are very different from values previously measured by μ-PL or PLE experiments in other InAs/InP QDs [8] and QSs [6, 9], where the biexciton binding energies and the energy spacing between the ground and first excited state ranged from 1 to 2 meV and from 15 to 30 meV, respectively. This confirms the growth of small QDs in this sample, unlike the quantum sticks usually observed [2–4] in this material system.

The μ-PL studies on other single QDs in the same sample have revealed the presence of several peaks even at low power density (figure 4(a) at 0.2 μW). Such a complex spectrum may be explained by the presence of charged excitons. Figure 4(a) shows the s-shell of a single QD as a function of the power density. At low excitation power three peaks are observed: a broad peak labelled c and two peaks at higher energy labelled a and b. When the excitation power is increased, the integrated PL intensity of these peaks increases linearly with the power. These peaks are therefore related to the double charged exciton, X^{2-} following previous results obtained by Urbaszek [10]. When the excitation power density continues to increase, new peaks are emerging. The integrated PL intensity of the broad peak labelled d increases quadratically with the power density. Thus, the peak is associated with the double charged biexciton $2X^{2-}$. A previous study of the fine structure of the $2X^{2-}$ already showed the presence of three peaks split by hundreds of μeV [10]. The three peaks are not clearly resolved in the spectra of figure 4(a). So, we performed a polarized μ-PL experiment on this single QD in order to explain this behaviour. In figure 4(b), peak d is measured along two different polarization axes. The spectrum performed along the x-direction clearly shows the expected three peaks of the $2X^{2-}$ recombination labelled d1, d2 and d3. Three peaks are also observed for the spectra performed along the y-direction, but they are shifted by 200 μeV compared to the other polarization direction. Such a splitting has already been observed for exciton and biexciton recombinations of a single QD and has been related to an in-plane asymmetry of the QD [11]. The small splitting value explains why the three peaks are not resolved in the un-polarized spectra of figure 4(a).

These charged states give us the opportunity to measure the exchange parameters in this InAs/InP QD. As discussed by Warburton *et al* [12], the splitting between peak c and the doublet (a, b) is due to the two possible final states after the photon emission: a triplet state (peaks a and b) and a singlet state (peak c). The linewidth of peak c is around 1 meV.

Such a broadening of the singlet state can be interpreted as a fast relaxation of the electron from the P to the S state which introduces an energy uncertainty for the final state [13]. For the triplet state the two remaining electrons have a parallel spin orientation and thus a spin flip must occur to allow for the relaxation of one electron between the two states. This phenomenon is relatively slow and the linewidth of the triplet is not broadened.

The splitting between singlet and triplet states is equal to twice the electron–electron exchange energy X_{sp}^{ee} where the exchange interaction energy is between an electron in the s state and another one in the p state. The splitting between peaks a and b is associated with the electron–hole exchange energy X_{pxs}^{eh}. The measured values in this QD are $X_{sp}^{ee} = 4.2$ meV and $X_{pxs}^{eh} = 0.6$ meV. The S–P electron exchange energy X_{sp}^{ee} is strong as compared to the values found for other QDs. For instance, values around 1.5–3.5 meV have been found for In(Ga)As/GaAs QDs emitting in the 0.9–1.05 μm range [10, 13, 14]. The large value of X_{sp}^{ee} measured in our sample can be explained by a stronger spatial confinement found in these InAs/InP QDs. The exchange energy X_{pxs}^{eh} is also relatively large as compared with other reported values which are around 0.2 meV [10]. The exchange energy X_{pxs}^{eh} is equal to the exchange energy which splits the radiative and non-radiative exciton ground states [10]. In QDs, this exchange energy increases strongly when the QD size is decreased [11] and is mainly linked with the QD volume. Such a large exchange energy (0.6 meV) further confirms the small size of the QDs under study.

Weakly charged dots have already been observed in the literature when QDs are grown on n-doped substrates: for example in InP/GaInP QDs [15]. Our sample is grown on a semi-insulating substrate. The charging of some dots could be explained by the presence of impurities or defects near these dots. These defects are tentatively attributed to phosphorus vacancies in InP layers, already observed in InAs/InP QSs [2, 6]. These vacancies will create an excess of electrons in dots surrounding the defect. The presence of both charged and neutral dots in the same sample suggests that the density of such defects is lower than the density of dots.

4. Conclusion

Neutral and charged multiexcitonic states are observed in single InAs/InP QDs, which were grown under specific MBE growth conditions, chosen in order to better control the shape and density of the self-organized InAs quantum islands grown by solid source MBE on InP(001). μ-PL spectroscopy allows the quantitative determination of a high value of energy spacing between the ground state and first excited state (90 meV) in such InAs/InP QDs. A large biexciton binding energy (about 4 meV) is also measured in these QDs, which also reveals a strong spatial confinement in these InAs/InP QDs. The exchange energy values also confirm the small QD size. Finally, the observation of both charged and neutral excitons from different single QDs in the same sample shows that defects or processes responsible for the QD charging are in relatively low density.

Acknowledgments

The authors wish to thank Taha Benyattou for fruitful discussions. This work was supported by the French ministry of research and technology (AC Nanoscience NanoQUB).

References

[1] Imamoglu A and Yamamoto Y 1994 *Phys. Rev. Lett.* **72** 210
[2] Gendry M *et al* 2004 *J. Appl. Phys.* **95** 4761
[3] Walther C, Hoerstel W, Niehus H, Erxmeyer J and Masselink W T 2000 *J. Cryst. Growth* **209** 572
[4] Dery H, Benisty E, Epstein A, Alizon R, Mikhelashvili V, Eisenstein G, Schwertberger R, Gold D, Reithmaier J P and Forchel A 2004 *J. Appl. Phys.* **95** 6103
[5] Dupuy E *et al* 2006 *Appl. Phys. Lett.* to be submitted
[6] Salem B, Benyattou T, Guillot G, Bru-Chevallier C, Bremond G, Monat C, Hollinger G and Gendry M 2002 *Phys. Rev.* B **66** 193305
[7] Cornet C, Even J and Loualiche S 2005 *Phys. Lett.* A **344** 457
[8] Chithrani D, Williams R L, Lefebvre J, Poole P J and Aers G C 2004 *Appl. Phys. Lett.* **84** 978
[9] Salem B, Chauvin N, Benyattou T, Guillot G, Bru-Chevallier C, Bremond G, Monat C, Rojo-Romeo P and Gendry M 2005 *Nanotechnology* **16** 444
[10] Urbaszek B, Warburton R J, Karrai K, Gerardot B D, Petroff P M and Garcia J M 2003 *Phys. Rev. Lett.* **90** 247403
[11] Bayer M *et al* 2002 *Phys. Rev.* B **65** 195315
[12] Warburton R J, Schäflein C, Haft D, Bickel F, Lorke A, Karrai K, Garcia J M, Schoenfeld W and Petroff P M 2000 *Nature* **405** 926
[13] Urbaszek B, McGhee E J, Krüger M, Warburton R J, Karrai K, Amand T, Gerardot B D, Petroff P M and Garcia J M 2004 *Phys. Rev.* B **69** 035304
[14] Finley J J *et al* 2001 *Phys. Rev.* B **63** 161305(R)
[15] Pistol M-E 2004 *J. Phys.: Condens. Matter* **16** S3737